Rapid Radiochemical Analysis of Radionuclides Difficult to Measure in Environmental and Waste Samples

Hou, Xiaolin; Shi, Keliang; Qiao, Jixin

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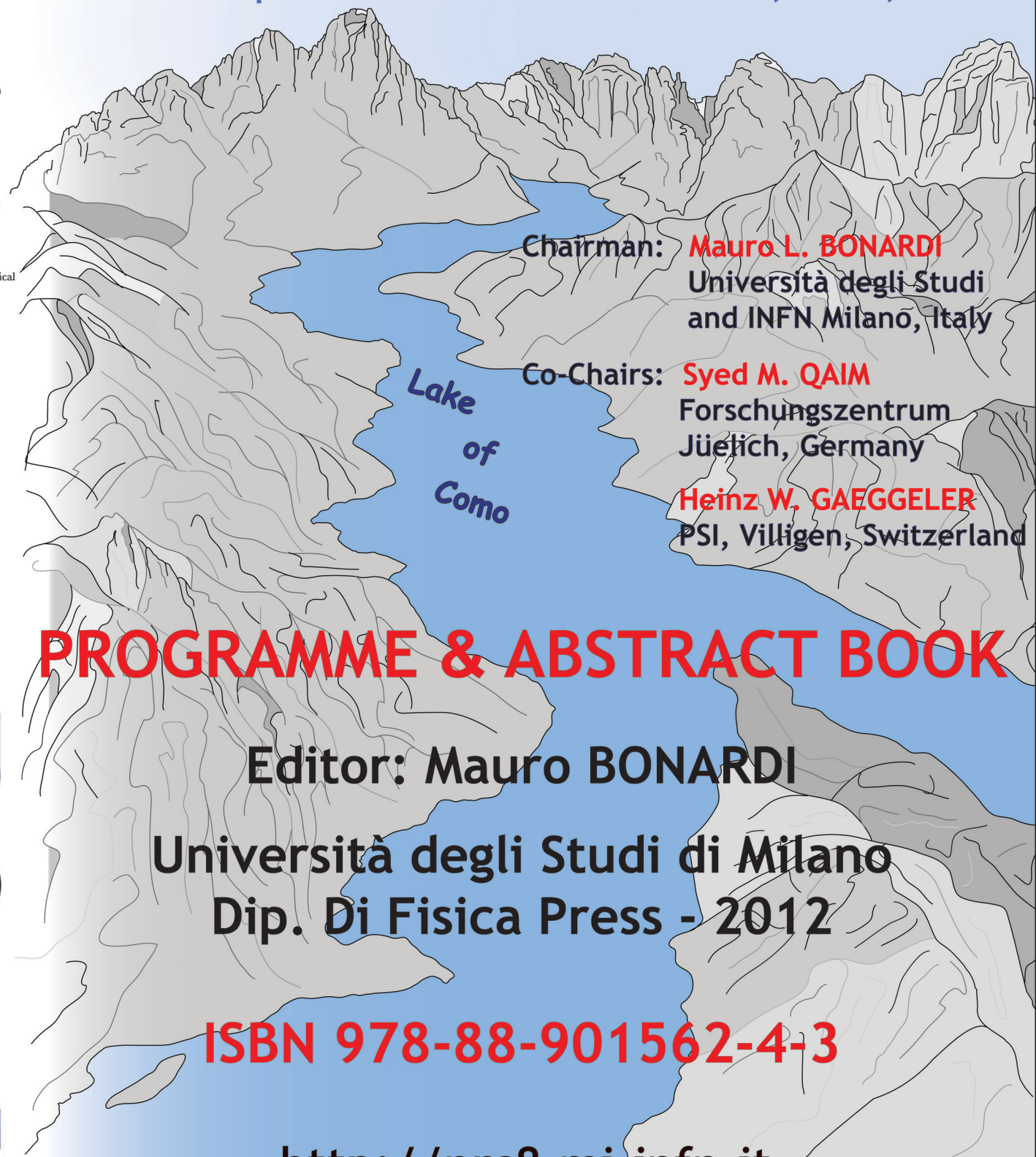

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PSI, Villigen, Switzerland

PROGRAMME & ABSTRACT BOOK

Editor: Mauro BONARDI

Università degli Studi di Milano
Dip. Di Fisica Press - 2012

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&

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NRC-8, EuCheMS International Conference on Nuclear and Radiochemistry

Sunday 16 September 2012 - Friday 21 September 2012

Como, Italy

Programme

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Sunday 16 September 2012

Reception - (16:00-18:00)

Registration at registration desk of Grand Hotel di Como

Lectio Magistralis - (18:00-19:00)

time	[id] title	presenter
18:00	[0] LECTIO MAGISTRALIS - Chemistry in Italy during the late 18^{th} and 19^{th} Centuries	Prof. BELLOBONO, Ignazio Renato

Welcome party: Reception cocktail in the "Grand Hotel di Como" garden - (19:30-21:00)

Monday 17 September 2012

Opening Ceremony - (08:00-09:30)

Opening Ceremony and Registration at registration desk of Grand Hotel di Como

time	[id] title	presenter
08:00	[263] A warm welcome from the Division of Nuclear and Radiochemistry (DNRC) of EuCheMS	Prof. GAEGGELER, Heinz (Paul Scherrer Institut, Switzerland)

Coffee break - (09:30-10:00)

Session 1 - Radiopharmaceutical Chemistry (radiodiagnostics, radiotherapy, theragnostics) - (10:00-13:00)

- Conveners: Prof. Cutler, Cathy (University of Missouri, USA)

time	[id] title	presenter
10:00	[250] OPENING LECTURE - Future prospects in diagnostic and therapeutic nuclear medicine	Prof. CHATAL, Jean-François (GIP ARRONAX, France)
10:30	[247] INVITED LECTURE - A Bridge not too Far: Personalized Medicine with the use of Theragnostic Radiopharmaceuticals	Prof. SRIVASTAVA, Suresh (Brookhaven National Laboratory, USA)
11:00	[171] INVITED LECTURE - On the road from Radiopharmacy to Molecular Imaging: the fundamental role of Technetium and Rhenium Chemistry	Prof. ALBERTO, Roger (University of Zurich, Switzerland)
11:20	[234] INVITED LECTURE - A renaissance of radionuclide generators for versatile application	Prof. ROESCH, Frank (University of Mainz, Germany)
11:40	[233] INVITED LECTURE - ^{90}Y and ^{177}Lu labelled peptides for PRRT: nuclear and radiochemical aspects	Dr. CHINOL, Marco (European Institute of Oncology, Italy) Dr. PAPI, Stefano (European Institute of Oncology, Italy)
12:00	[96] ORAL PRESENTATION - Radiopharmacological studies of leptin and thyroid hormones relationship in white adipose tissue	Dr. PAVELKA, Stanislav (Department of Radiometry, Institute of Physiology, v.v.i., Academy of Sciences of the Czech Republic, Prague; and Institute of Biochemistry, Masaryk University, Brno, Czech Republic)
12:15	[20] Testing the feasibility of 44mSc/44Sc as a potential in vivo generator for PET imaging and an alternative to the existing 44Ti/44Sc ?	Dr. HUCLIER-MARKAI, Sandrine (Laboratoire Subatech, UMR 6457, Ecole des Mines de Nantes /CNRS/IN2P3 / Université de Nantes, 4 Rue A. Kastler, BP 20722, F-44307 Nantes Cedex 3, France.)
12:30	[136] ORAL PRESENTATION - ^{44,43}Sc and ^{47}Sc as matched pair for theranostic approach to peptide receptor radionuclide therapy	Mr. KRAJEWSKI, Seweryn (Institute of Nuclear Chemistry and Technology, Dorodna Street 16, 01312 Warsaw, Poland)
12:45	[152] ORAL PRESENTATION - TiO_2 nanoparticles as vehicles of ^{212}Pb and ^{225}Ac for internal radiotherapy	Ms. LESZCZUK, Edyta (Institute of Nuclear Chemistry and Technology, Poland)

Lunch - (13:00-14:00)

Session 2 (cn't of Session 1) - Radiopharmaceutical Chemistry (radiodiagnostics, radiotherapy, theragnostics) - (14:00-17:00)

- Conveners: Prof. Chatal, Jean-Francois (GIP ARRONAX, France); Dr. Chinol, Marco (European Institute of Oncology, Italy)

time	[id] title	presenter
14:00	[255] OPENING LECTURE - Organic PET-Radiopharmaceuticals – Aspects of Previous and Current Labelling Techniques	Prof. STEINBACH, Jörg (Helmholtz-Zentrum Dresden-Rossendorf, Germany)
14:20	[89] INVITED LECTURE - Novel ^{18}F-Radiochemistry	Dr. ERMERT, Johannes (Forschungszentrum Juelich GmbH, Institut fuer Neurowissenschaften und Medizin, INM-5: Nuklearchemie, Germany)
14:40	[163] INVITED LECTURE - Labeling of radiopharmaceuticals with Iodine-124 and their clinical applications	Dr. PILLARSETTY, Naga Vara Kishore (Memorial Sloan-Kettering Cancer Center, USA)
15:00	[22] INVITED LECTURE - Recoil and conversion electron implications to be taken into account in the design of therapeutic radiopharmaceuticals utilising in vivo generators	Prof. ZEEVAART, Jan Rijn (Necsa, South Africa)
15:20	[239] INVITED LECTURE - Radiochemistry of Astatine-211: Application to Alpha Particle Targeted Radiotherapeutics	Prof. ZALUTSKY, Michael (Duke University, USA)
15:40	[46] ORAL PRESENTATION - Polymersomes as nano-carriers for alpha radionuclide therapy	Dr. DENKOVA, Antonia (TU Delft, Netherlands)
15:55	[192] ORAL PRESENTATION - Production of Four Terbium Radioisotopes for Radiopharmaceutical Applications	Mr. DORRER, Holger (Paul Scherrer Institut, Villigen-PSI, Switzerland & University of Bern, Berne, Switzerland)
16:10	[160] ORAL PRESENTATION - Decay data measurements on ^{213}Bi using recoil atoms	Dr. POMMÉ, Stefaan (EC-JRC-IRMM, Belgium)
16:25	[149] ORAL PRESENTATION - Bioconjugated nanozeolites labeled with ^{223,224,225}Ra	Prof. BILEWICZ, Aleksander (Institute of Nuclear Chemistry and Technology, Poland)
16:40	[179] ORAL PRESENTATION - Development of ^{44}Sc production for radiopharmaceutical applications	Mrs. BUNKA, Maruta (Laboratory of Radiochemistry and Environmental Chemistry, Villigen PSI, Switzerland)

Coffee breack - (17:00-17:30)

Poster Session: 1, Session dedicated to the 80th Birthday of Prof. (em) Ignazio Renato Bellobono - (17:30-19:00)

[id] title	presenter	board
[138] Electrosynthesis of electrophilic n.c.a. 18F-fluorinating reagents	Mr. DRERUP, Christian	
[140] Gallium-68 complexes of NOTA-bis(phosphonates) conjugates as radiotracers for bone imaging with PET	Mr. HOLUB, Jan	

[217] Radiosynthesis of 18F-Labeled Diclofenac Hydroxy-Derivative as potential micro-PET imaging tracer	LIN, Manjing	
[97] Quantification of iodothyronine deiodinases activities, induced in cultured astrocytes by purinergic agonists, with the use of radiometric enzyme assays	Dr. PAVELKA, Stanislav	
[130] On the way to the synthesis of the first transactinide carbonyl complex	Dr. EVEN, Julia	
[126] Liquid-phase Studies of Seaborgium using the Automated Liquid-liquid Extraction system SISAK	Dr. OMTVEDT, Jon Petter	
[24] Gas-phase chemistry of carbonyl complexes formed in hot-atom reactions with short-lived isotopes of a Cf-252 fission source	WANG, Yang Prof. QIN, Zhi	
[36] Characterization of At- and AtO+ species in simple media by high performance ion exchange chromatography coupled to gamma detector. Application to astatine speciation in human serum.	Dr. SABATIÉ-GOGOVA, Andrea	
[86] Redox studies of the heaviest elements using an electrolytic column apparatus	Dr. TOYOSHIMA, Atsushi	
[74] On the fast release of tracer elements from metallic hosts – a step towards vacuum chromatography	Dr. EICHLER, Robert	
[197] Automated rapid α/SF detection system for studying aqueous chemistry of superheavy elements at RIKEN	Dr. HABA, Hiromitsu	
[164] Extraction of Zr and Hf using TBP and TIOA for the chemistry of element 104 Rf	Dr. KASAMATSU, Yoshitaka	
[202] Development of a liquid scintillation detection system for aqueous chemistry of seaborgium	Mrs. KOMORI, Yukiko	
[158] Diamond Detectors in Transactinide Chemistry	Mr. STEINEGGER, Patrick	
[200] Extraction behavior of Mo(VI), Mo(V), W(VI), and W(V) from HCl solutions by Aliquat 336	Mr. YOKOKITA, Takuya	
[61] Study on Quadrivalent Chemical Species of Rutherfordium in Aqueous Solution by Means of TTA resin	Dr. YOKOYAMA, Akihiko	
[211] Characteristics of Uranium species when U(III) in a LiCl-KCl molten salt was leached out with water and ionic liquid	Dr. IM, Hee-Jung	
[55] Selective ion exchangers for Fukushima waste effluent purification	Prof. LEHTO, Jukka	
[21] Experimental Investigation on Cryogenic Adsorption of Low-concentration Hydrogen from Helium by MS5A	Ms. QIAN, xiaojing	
[139] The Effect of Cellulose Degradation Products on the Migration of 90Sr in Cementitious Backfill using Radial Diffusion.	Mr. HINCHLIFF, John	
[110] Recoil-radiolabelling of nanoparticles with 7Be generated by 7Li(p,n)7Be reaction in mixed powder targets.	Dr. KOZEMPEL, Jan	
[53] Study of the Production of Mo and Tc Medical Radioisotopes Via Proton Induced Nuclear Reaction on natMo.	Dr. ALHARBI, Abeer	
[162] Synthesis and Characterization of Radiolabelled Silver Nanoparticles	Dr. ICHEDEF, Cigdem	
[1] Nuclear and radioanalytical techniques in nanotoxicology research: studies on the Rabbit Reproductive System	Prof. GROPPI, Flavia	
[204] Synthesis of new 18F-labelled Porphyrins and their potential aplication for in vivo Molecular Imaging with PET	Ms. SIMÕES, Ana	
[155] New analytical method for actinide (Pu, Am, U, Th, Np) separation based on diglycolamide resin (DGA)	Ms. GROSKA, Judit	

[47] Sequential separation and determination of Pu, Sr-90 and Am-241 in soil and sediment samples using DGA Resin for the preconcentration of the actinides.	Dr. JÄGGI, Maya	
[187] Neptunium Redox Chemistry in Irradiated HNO3 Solutions	Dr. PAULENOVA, Alena	
[125] Determination of Boron distribution in Co-Re alloys	Dr. SZENTMIKLÓSI, László	
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[105] Radiochemical separation of arsenic from selenium and its potential usage in generator isotope production.	Dr. CHAJDUK, Ewelina	
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[214] A new PTS for short-time neutron activation analysis	Prof. ISMAIL, Saleh	
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[132] Detection of irradiated foods using TL, ESR and GC/MS	Mrs. KANG, Yoonjung	
[189] The air-water partitioning of radon in groundwater contaminated by BTEX	Dr. LEE, Kil Yong	
[157] Geochemically anomalous phonolites from Lusatian Mountains, Czech Republic: Possible source materials and processes of their origin	Dr. MIZERA, Jiří	
[131] Determination of Mineral Contents in Korean Domestic Unpolished Rice and Bean Samples by Neutron Activation Analysis	Mr. MOON, JongHwa	
[166] Non-destructive and quantitative multi-elemental analysis by muonic X-ray spectroscopy for archeological bronze samples	Dr. NINOMIYA, Kazuhiko	
[98] Radiometric quantification of type 1 iodothyronine 5'-deiodinase activity in human white adipose tissue	Dr. PAVELKA, Stanislav	
[99] Development of diet-induced obesity in the rat, followed by radioanalytical methods	Dr. PAVELKA, Stanislav	
[186] Mineral Elements Determination in Medicinal Plants	SILVA, Paulo	
[183] Validation of the method for Ni determination in NPP evaporator concentrates	Dr. SULAKOVA, Jana	
[168] A preliminary study of prompt gamma-ray activation analysis using pulsed neutron at J-PARC / ANNRI	Dr. TOH, Yosuke	
[51] Biodistribution of Gadolinium-Based Contrast Agent, and Concentration of Trace Elements in Normal and Nephrectomized Mice	Mr. WASHIYAMA, Kohshin	
[195] Application of medium-energy proton beam from AIC-144 cyclotron in biological and environmental studies	Ms. WÓJCIK, Anna	
[43] Sequential separation of ultra-trace U, Th, Pb, and lanthanides with a single anion-exchange column	Dr. MIYAMOTO, Yutaka	
[172] Experimental Performance Evaluation of a Compton Suppression System for Neutron Activation Analysis by Using a Gamma-ray Source and Standard Reference Materials	Mr. MOON, JongHwa	
[123] NukWik – A Tool for Collaboration and Sharing Teaching Material in Radiochemistry	Mr. NORÉN, Henrik Mr. LERUM, Hans V.	

[48] Fluorescent Imaging of the Radiation Dose Surrounding an Iridium-192 Seed Used in Brachytherapy	Dr. DENKOVA, Antonia	
[193] 66Ga-labeling of DOTA-conjugated cyclic RGDfK dimer for αvβ3 integrin overexpression tumors	AVILA-RODRIGUEZ, Miguel	
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[258] SYNTHESIS AND X-RAY STUDY OF RADIUM METAPLUMBATE	BUTKALYUK, Irina	
[261] The influence of iron on the efficiency of the 68Ga labeling of DOTATOC and simple colorimetric determination of iron	Dr. MUELLER, Dirk	
[262] Separation of actinium-225 for nuclear medicine purposes from thorium targets irradiated by high energy protons	ALIEV, Ramiz	
[59] Fast beta-alpha-pile-up suppression electronics for super heavy element identification	Dr. DRESSLER, Rugard	
[143] Advanced Fuels for Generation IV reactors: Reprocessing and Dissolution – ASGARD	Dr. RETEGAN, Teodora	
[268] Problems of determination of Tc-99 in soil and sediments	Mr. KLESZCZ, Krzysztof Prof. MIETELSKI, Jerzy	

Tuesday 18 September 2012

Session 3 - Chemistry of radioelements and Super Heavy Elements research - (08:00-11:30)

- Conveners: Prof. Gaeggeler, Heinz (PSI, Switzerland); Dr. Schaedel, Matthias (Japan Atomic Energy Agency, GSI, Germany)

time	[id] title	presenter
08:00	[178] OPENING LECTURE - Recent Advances in Superheavy Element Research	Prof. TÜRLER, Andreas (Laboratory of Radiochemistry and Environmental Chemistry, Paul Scherrer Institute & Bern University, Switzerland)
08:30	[241] INVITED LECTURE - Spectroscopic methods for the heaviest nuclei	Prof. HERZBERG, Rolf-Dietmar (University of Liverpool, UK)
09:00	[63] INVITED LECTURE - The unique chemical and physical properties of the heaviest elements in the Periodic Table	Prof. KRATZ, Jens Volker (Universität Mainz, Germany)
09:30	[169] INVITED LECTURE - Vacuum thermochromatography - prospective method for heaviest element studies	Prof. ZVARA, Ivo (JINR Dubna, Russia)
09:50	[111] INVITED LECTURE - Aqueous-phase chemistry of the heaviest elements	Dr. NAGAME, Yuichiro (Japan Atomic Energy Agency, Japan)
10:10	[76] ORAL PRESENTATION - The observation of a volatile compound formation with Po and Bi during experiments with superheavy elements	Dr. EICHLER, Robert (Paul Scherrer Institute,Switzerland)
10:25	[146] ORAL PRESENTATION - New experiments to study properties of ^{268}Db produced in the ^{48}Ca + ^{243}Am reaction	Dr. AKSENOV, Nikolay (Flerov Laboratory of Nuclear Reactions, Joint Institute for Nuclear Research, Russia)
10:40	[71] ORAL PRESENTATION - Superheavy element 114 is a volatile metal	Prof. DÜLLMANN, Christoph (University of Mainz + GSI Darmstadt + Helmholtz Institute Mainz, Germany)
10:55	[35] ORAL PRESENTATION - Exploration of the metallic character of astatine	MONTAVON, Gilles (Subatech, France)
11:10	[207] ORAL PRESENTATION - RIKEN GARIS as a promising interface for superheavy element chemistry –Production of 261Rf, 262Db, and 265Sg for chemical studies using the GARIS gas-jet system–	Dr. HABA, Hiromitsu (RIKEN, Japan)

Coffee break - (11:30-11:50)

Session 4 - Reaction mechanisms and nuclear recoils, nuclear base spectroscopies, radiation geochronology, isotope effects - (11:50-13:30)

- Conveners: Dr. Schumann, Dorothea (PSI, Switzerland); Prof. Revay, Zsolt (Technical University Munich, Geramany)

time	[id] title	presenter
11:50	[196] OPENING LECTURE - Iron speciation in aqueous systems: the power of Mössbauer spectroscopy applied in frozen solutions	Prof. HOMONNAY, Zoltan (Eotvos Lorand University, Hungary)
12:10	[253] INVITED LECTURE - Half lives of nuclides for geological use: 2012 evaluations for ^{87}Rb, ^{235}U and ^{234}U	Prof. VILLA, Igor M (Universität Bern, Switzerland; Università Milano Bicocca, Italy)

12:30	[222] INVITED LECTURE - Rapid Radiochemical Analysis of Radionuclides Difficult to Measure in Environmental and Waste Samples	Dr. HOU, Xiaolin (Center for Nuclear Technologies, Technical University of Denmark, Denmark)
12:50	[72] INVITED LECTURE - TRIGA-SPEC: an apparatus for high-precision mass spectrometry and laser spectroscopy on short-lived neutron-rich radionuclides produced at the research reactor TRIGA Mainz	Mr. SMORRA, Christian (Max-Planck-Institut für Kernphysik Heidelberg, Ruprecht Karls-Universität Heidelberg, Johannes Gutenberg-Universität Mainz, Germany)
13:10	[201] ORAL PRESENTATION - Local Fields at Nonmagnetic Probe Sites in a Perovskite La0.7Ca0.3MnO3	Dr. SATO, Wataru (Institute of Science and Engineering, Kanazawa University, Japan)

Lunch - (13:30-14:30)

Session 5 - Nuclear fuel cycles, Research Reactors and present NPP (including Gen IV and Th reactors) - (14:30-16:30)

- Conveners: Prof. Geckeis, Horst (Karlsruhe Institute of Technology, Germany); Dr. Saracco, Paolo Giovanni (INFN Genova, Italy)

time	[id] title	presenter
14:55	[235] INVITED LECTURE - Update of GEN-IV reactors and lead cooled reactors	CINOTTI, Luciano (M.E.Rivus s.r.l, Italy)
15:30	[165] INVITED LECTURE - Nuclear energy chemistry and recent progresses in nuclear fuel reprocessing in China	Dr. SHI, Wei-Qun (Institute of High Energy Physics,Chinese Academy of Sciences, China)
15:50	[259] INVITED LECTURE - Nuclear Fuel Cycle: Processes, critical aspects and perspectives	Dr. TROIANI, Francesco (ENEA/NUCLECO SpA) Dr. GRASSO, Giacomo (ENEA)
16:10	[252] INVITED LECTURE - Solution reactors for production of Mo-99 and Sr-89 (via Kr-89)	Dr. PAVSHUK, Vladimir A. (Russian Research Center “Kurchatov Institute”, Russia)

Coffee break - (16:30-16:50)

Session 6 (cn't of Session 5) - Nuclear fuel cycles, Research Reactors and present NPP (including Gen IV and Th reactors) - (16:50-19:30)

- Conveners: Prof. Pagani, Carlo (University of Milano and INFN, Italy); Prof. Aggarwal, Suresh Kumar (Bhabha Atomic Research Centre, India)

time	[id] title	presenter
16:50	[231] INVITED LECTURE - Conceptual design of a low power ADS with a 70 MeV proton beam for reasearch and training	Dr. SARACCO, Paolo Giovanni (Istituto Nazionale di Fisica Nucleare, Sez. Genova, Italy)
17:10	[236] INVITED LECTURE - High power superconducting proton accelerators for ADS and Gen-IV	Prof. PAGANI, Carlo (University of Milano and INFN LASA, Italy)
17:30	[134] INVITED LECTURE - Transmutation of minor actinides in the molten salt reactor recently studied in Russia	Prof. PONOMAREV, Leonid (NRC "Kurchatov Institute", Russia)
17:50	[145] INVITED LECTURE - The very powerful UCN source at the reactor TRIGA Mainz - Application for precise measurements of the neutron half-life	Dr. SOBOLEV, Yury (Johannes-Gutenberg Universität Mainz, Institut für Kernchemie, Germany)

18:10	[151] ORAL PRESENTATION - Adsorption of selected fission products on various forms of TiO_2 nanoparticles.	Ms. FILIPOWICZ, Barbara (Institute of Nuclear Chemistry and Technology, Dorodna 16, 03-195 Warsaw, Poland)
18:25	[40] ORAL PRESENTATION - Development of Decontamination Method Using Ionic Liquid as a Medium for Treating Waste Contaminated with Uranium	Mr. OHASHI, Yusuke (Ningyo-toge Environmental Engineering Center, Japan Atomic Energy Agency, Japan)
18:40	[137] ORAL PRESENTATION - Selectivity of bis-triazinyl bipyridine ligands for americium(III) in Am/Eu separation by solvent extraction. Quantum mechanical study	Prof. NARBUTT, Jerzy (Institute of Nuclear Chemistry and Technology, Warsaw, Poland)
18:55	[210] ORAL PRESENTATION - Decomposition of boric acid solutions and evolution of gases under mixed thermal and fast neutrons and gamma radiation	Dr. IM, Hee-Jung (Nuclear Chemistry Research Division, Korea Atomic Energy Research Institute, Korea)

Wednesday 19 September 2012

Session 7 - Nuclear Chemistry, Radionuclide Production, High-Power Targetry - (08:00-10:45)

- Conveners: Prof. Zhuikov, Boris (Institute for Nuclear Research of Russian Academy of Sciences, Russia); Prof. Bonardi, Mauro L. (UNIMI and INFN - Milano, Italy)

time	[id] title	presenter
08:00	[120] OPENING LECTURE - Recent advances in nuclear data research for medical radionuclide production	Prof. QAIM, Syed M. (Forschungszentrum Jülich, Germany)
08:30	[127] INVITED LECTURE - ^{68}Ge-^{68}Ga production revisited: new excitation functions, target preparation and separation chemistry	Prof. HERMANNE, Alex (Cyclotron lab, Vrije Universiteit Brussel, Belgium)
08:50	[243] INVITED LECTURE - Radionuclide production studies by heavy ion beams	Prof. LAHIRI, Susanta (Saha Institute of Nuclear Physics, India)
09:10	[256] Overview of PET radionuclide production methods.	Prof. CLARK, John C (University of Edinburgh, UK)
09:30	[254] INVITED LECTURE - Radionuclides and radiopharmaceuticals at POLATOM	Dr. MIKOLAJCZAK, Renata (NCBJ Radioisotope Centre POLATOM, Poland)
09:50	[240] INVITED LECTURE - Reactor production of radionuclides for molecular imaging and targeted radiotherapy	Dr. CUTLER, Cathy (University of Missouri, USA)
10:10	[260] INVITED LECTURE - Review of Mo-99 and other reactor radionuclide production in RIAR in terms of world demands	Dr. KUZNETSOV, Rostislav (Research Institute of Atomic Reactors, Russia)
10:30	[41] ORAL PRESENTATION - A new route for polonium-210 production from a bismuth-209 target	Mr. YOUNES, Ali (SUBATECH laboratory (UMR 6457), Nantes 44307, France)

Coffee break - (10:45-11:00)

Session 8 - Nuclear Chemistry, Radionuclide Production, High-Power Targetry - (11:00-13:30)

- Conveners: Prof. Srivastava, Suresh (Brookhaven National Laboratory, USA); Dr. Mikolajczak, Renata (Polatom, Poland)

time	[id] title	presenter
11:00	[245] OPENING LECTURE - The Road to Cyclotron Produced Tc-99m	Prof. MCQUARRIE, Steve (University of Alberta, Canada)
11:20	[32] INVITED LECTURE - Cyclotron production of radionuclides with medium-energy proton beams and high-power targetry	Dr. STEYN, G. (iThemba LABS, South Africa)
11:40	[29] INVITED LECTURE - Radionuclide Production at Accelerator with High Power Targets	Dr. ZHUIKOV, Boris (Institute for Nuclear Research of Russian Academy of Sciences, Russia)
12:00	[242] INVITED LECTURE - Recent advances in large scale isotope production at LANL	Dr. NORTIER, Francois (Los Alamos National Laboratory, USA)
12:20	[237] INVITED LECTURE - ARRONAX: on the way to the production of radio-isotopes with high-power targets	Dr. HADDAD, Ferid (Subatech / ARRONAX, France)
12:40	[238] INVITED LECTURE - Use of radioactive targets for production of therapy radionuclides at the Brookhaven Linac Isotope Producer	Dr. MAUSNER, Leonard (Brookhaven National Laboratory, USA)

13:00	[124] ORAL PRESENTATION - Thermochromatography study of volatile Tellurium species in various gas atmospheres.	Dr. MAUGERI, Emilio Andrea (PSI, Switzerland)
13:15	[107] ORAL PRESENTATION - Complexation of Cm(III) with 2,6-bis(5-(2,2-dimethylpropyl)-1H-pyrazol-3-yl)pyridine (C5-BPP) studied by time-resolved laser fluorescence spectroscopy	BREMER, Antje (Institute for Nuclear Waste Disposal - Karlsruhe Institute of Technology, Geramany)

Lunch - (13:30-14:30)

Celebration of 50th Birthday of Radiochimica Acta - (14:30-14:50)

- Conveners: Prof. Qaim, Syed M. (Forschungszentrum Jülich); Prof. Kratz, Jens Volker (University of Mainz); Sperlich, Angelika (Oldenbourg Verlag)

Session 9 - Applications of radiotracers and nanoparticles - (14:50-17:40)

- Conveners: Prof. Chatt, Amares (Dalhousie University, Canada); Prof. Hou, Xiaolin (Technical University of Denmark, Denmark)

time	[id] title	presenter
14:50	[78] OPENING LECTURE - Nuclear and radioanalytical techniques in nanotoxicology research	Dr. SABBIONI, Enrico (European Center for the Sustainable Impact of Nanotechnology-ECSIN, Veneto Nanotech ScpA, Italy)
15:10	[227] INVITED LECTURE - Radiochemical neutron activation analysis: the continuous need of this analysis mode	Prof. KUCERA, Jan (Nuclear Physics Institute, AS CR, CZ-25068 Husinec-Rez 130, Czech Republic)
15:30	[82] INVITED LECTURE - Nuclear analytical methods in prostate cancer diagnostics	Dr. ZAICHICK, Vladimir (Medical Radiological Research Center, Russia)
15:50	[246] INVITED LECTURE - The use of nuclear analytical techniques in the identification and investigation of metal- and metalloid-containing proteins	Prof. BEHNE, Dietrich (Helmholtz Centre Berlin, Germany)
16:10	[56] INVITED LECTURE - Exploitation of accelerator waste for the production of exotic radionuclides	Dr. SCHUMANN, Dorothea (Paul Scherrer Institute, Switzerland)
16:30	[185] INVITED LECTURE - Novel mesoporous materials for actinide and lanthanide separation	Prof. NITSCHE, Heino (University of California, Berkeley, Department of Chemistry, USA)
16:50	[150] ORAL PRESENTATION - Radiolabelling of engineered nanoparticles – different strategies for Ag^0-NP, TiO_2-NP and MWCNT	Dr. FRANKE, Karsten (HZDR, Germany)
17:05	[52] ORAL PRESENTATION - Synthesis of Silica-coated Bimetallic Nanoparticles as Radiotracers	Dr. JUNG, Sung-Hee (Korea Atomic Energy Research Institute, Korea)
17:20	[42] ORAL PRESENTATION - Nanomedicine Approaches of Radioactive Gold Nanoparticles In Cancer Therapy	Prof. KATTI, Kattesh (University of Missouri, USA)

Coffee break - Offered by Oldenburg-Verlag - (17:40-18:00)

Poster Session: 2, Session dedicated to Prof. Attila Vertes (deceased on 31 December 2011) - (18:00-19:50)

[id] title	presenter	board

[69] The LARAMED project at INFN Legnaro National labs	Dr. ESPOSITO, Juan	
[118] Validation of neutron induced data up to 18 MeV for production of the therapeutic radionuclide 67Cu	Dr. HUSSAIN, Mazhar	
[67] Radiochemical separation of no-carrier-added 97Ru and 95Tc produced by 12C-induced reaction on natural yttrium target	Prof. LAHIRI, Susanta	
[91] A novel path in partitioning: Water-soluble BTP ligands for the innovative SANEX process	Prof. PANAK, Petra	
[94] Pd-based intermetallic targets for high intensity irradiations	Mr. USOLTSEV, Ilya	
[79] Development of actinide liquid scintillating target	Dr. AUPIAIS, Jean	
[70] Preparation of TBq Activity 7Be from SINQ Cooling Water	Mrs. STOWASSER, Tanja	
[147] Development of production possibilities of n.c.a radiomanganese in a non aggressive and toxic medium	Dr. BUCHHOLZ, Martin	
[85] Separation of carrier free 177Lu from 177Lu /Yb mixture by electro-amalgamation of ytterbium	Dr. CIESZYKOWSKA, Izabela	
[93] Preparation of 57Co sources for Mössbauer Spectroscopy	Dr. ŻÓŁTOWSKA, Małgorzata	
[64] Accelerator-based Alternative Tc-99m production: EMPIRE 3.1 theoretical simulations of cross sections for Mo(p,X) reactions and comparison with literature experimental data	Dr. ESPOSITO, Juan	
[133] Cyclotron produced ^{45}Ti – production, purification and yields	Dr. FRANKE, Karsten	
[249] Separation of no-carrier-added 109Cd from natural silver target using RTIL 1-butyl-3-methylimidazolium hexafluorophosphate	Prof. LAHIRI, Susanta	
[81] Application of a mixed bed column for the removal of iodine from radioactive process effluents	Dr. HAPPEL, Steffen	
[39] 64Cu and 67Cu Production and Purification Research at the Radiation Science and Engineering Center at the Pennsylvania State University	Prof. UNLU, Kenan	
[156] Measurements of γ- and β+-intensities of Ti-45	Mr. KUHN, Sebastian	
[100] Production and purification of 56Co at the Leipzig cyclotron	Dr. MANSEL, Alexander Dr. FRANKE, Karsten	
[181] Separation of radioiodine by dry distillation process from irradiated elemental Te target	Dr. MISIAK, Ryszard Mr. BARTYZEL, Mirosław Mr. WĄS, Bogdan	
[148] An Automated Production of 64Cu on 18/9 MeV cyclotron	Prof. RAJEC, Pavol	
[208] Radiochemical and cross section studies for the production of the therapeutic radionuclide ^{193m}Pt	Dr. SCHOLTEN, Bernhard	
[119] Nuclear spallation reactions in chromium, yttrium and terbium with 386 MeV neutrons	Dr. SEKIMOTO, Shun	
[199] Nuclear and radiochemical study of production and utilization of radioactive astatine isotopes in the 7Li+natPb reaction	Dr. NISHINAKA, Ichiro	
[142] Polonium Evaporation Studies from Liquid Lead-based Alloys	Mr. RIZZI, Matthias	
[220] Elemental analysis of rivers, marshes and ground water in Thi Qar region, Iraq	Dr. ALRAKABI, Muhanad	
[221] Investigation of uranium contamination in ground water of southwest Punjab using EDXRF technique	Dr. ALRAKABI, Muhanad	

[50] Sorption of Cadmium, Nickel, Caesium and Strontium to a Laterite Soil: Application of Linear Additive Model and Surface Complexation Modelling	Mr. ANJOLAIYA, Olanrewaju	
[106] Interaction of Cm(III) with human serum transferrin studied by Time-Resolved Laser Fluorescence Spectroscopy (TRLFS)	BAUER, Nicole	
[62] Application of Non-linear Heterogeneity-based Isotherm Models for Charactering Sorption of Cs and Se on Mudrocks	Dr. CHUAN-PIN, Lee	
[87] Experiments to demonstrate chemical containment: Solubility under the cementitious conditions of a repository in the UK	Dr. FELIPE-SOTELO, Monica	
[109] Attachment of APTES ((3-aminopropyl)triethoxysilane) to silica for sorption and selective removal of radionickel from solution	Mr. HOLT, James	
[230] Chemical Decontamination, at Field, after 137Cs Accident at Goiania, Brazil	Prof. MEDEIROS, Joao Alfredo	
[225] Assessment of present and future radioactive contamination at global scale	Dr. NAVARRETE, Manuel	
[116] Trivalent Actinide/Lanthanide Sorption under Saline Conditions	Dr. RABUNG, Thomas	
[77] Structural studies of actinide-peptide complexes	Dr. SAFI, Samir	
[174] Iodine - 129 in water samples from Germany	Ms. SCHWINGER, Mareike	
[102] Archaeometry with INAA at the Research Reactor TRIGA Mainz	Mr. STIEGHORST, Christian	
[49] Characterisation of Radioactive Scales (NORM) Produced by the Onshore Oil and Gas Industry in the UK	Mr. AFOLABI, Oluwasola	
[129] Structural Transformations in Metallic Iron under the Action of External Irradiation	Prof. ALEKSEEV, Igor	
[114] THEREDA – a Thermodynamic Reference Database project	Dr. ALTMAIER, Marcus	
[103] Characterization of the Natural Organic Matter (NOM) by ultrafiltration and fluorescence in a groundwater plume contaminated with 60Co and 137Cs	Dr. CARON, François	
[190] Iodine-129 and iodine-127 in aerosols from Northern Germany	Dr. DARAOUI, Abdelouahed Ms. SCHWINGER, Mareike Dr. RIEBE, Beate	
[28] A Solid-State NMR Study of the Complexation of 109Cd with Isosaccharinic Acid	Dr. EVANS, Nick	
[88] Generation of inorganic colloids in the chemical disturbed zone in the proximity of a cementitious repository	Dr. FELIPE-SOTELO, Monica	
[219] Uranium in ground water samples from Anthemountas Basin, Northern Greece	Prof. IOANNIDOU, Alexandra	
[226] Time lag between the tropopause height and ^{7}Be activity concentrations on surface air	Prof. IOANNIDOU, Alexandra	
[209] Airborne radionuclides measured in Wako, Japan, after the Fukushima Dai-ichi nuclear power plant accident in 2011	Mr. KANAYA, Jumpei	
[26] Coprecipitation of Radionuclide Microquantities on Chitosans of Different Molecular Masses in Solutions	Prof. KULYUKHIN, Sergey	
[213] Environmental impact due to the operation of a tin and lead industry inferred by lichens	Dr. LEONARDO, Lucio	
[176] Spectroscopic Studies of Complexation Behaviour of Uranium(VI) by Schiff Bases	Ms. LINDNER, Katja	
[205] Preparation of spiked grass for use as environmental radioactivity calibration standard	Dr. LOURENÇO, Valerie	

[215] Gamma external radiation dose for Mexican population	Dr. NAVARRETE, Manuel	
[144] EC Interlaboratory Comparison on Radionuclides in Dried Bilberries	Dr. MEREŠOVÁ, Jana	
[167] Anomalous uranium enrichment in coals from Odeř, Sokolov Basin, Czech Republic	Dr. MIZERA, Jiří	
[68] Study of Uranium Behavior in Lignite Sediments from Ruprechtov Natural Analogue Site	Mr. PIDCHENKO, Ivan Dr. SUKSI, Juhani	
[112] Selective liquid-liquid extraction of Sr-85 with modified calixarenes	Ms. POETSCH, Maria Dr. MANSEL, Alexander	
[115] CROCK: Crystalline Rock Retention Processes A 7th Framework Programme Collaborative Project (2011-2013)	Dr. RABUNG, Thomas	
[182] Iodine-129 and iodine-127 in soils from Germany	Dr. RIEBE, Beate	
[212] Sorption of niobium on Olkiluoto soil samples	SÖDERLUND, Mervi	
[180] TiO2 based absorber for uranium separation and 236U measurement with AMS	Dr. NEMEC, Mojmir	
[84] Fractionation of U, Th, Ra and Pb from boreal forest soils by sequential extractions	Ms. VIRTANEN, Sinikka	
[117] Redox behavior of the Tc(VII)/Tc(IV) couple in dilute to concentrated NaCl and MgCl2 solutions	Mrs. YALCINTAS, Ezgi	
[83] Pu-240/Pu-239 atom ratios in the northern North Pacific and equatorial Pacific water columns	Prof. YAMADA, Masatoshi	
[224] Improvement of Detection Limits for Gamma-Ray Emitting Naturally Occurring Radionuclides in Drinking Water and Biological Materials by Instrumental Analysis using Compton Suppression Spectrometry	Prof. CHATT, Amares	
[191] Atmospheric activity concentration of radiocesium at Mikamine, Sendai and radioactivity distribution on the collection filters used in the measurement.	Dr. KIKUNAGA, Hidetoshi	
[198] Radioactivity measurement for air-dust and soil collected in eastern Japan area after the nuclear accident at the Fukushima Daiichi Nuclear Power Station	Mr. ZHANG, Zijian	
[27] The Thermal Decomposition of CH3131I in a Gas Phase	Prof. KULYKHIN, Sergey	
[66] Retardation behavior of Sr and Cs in Crushed and Intact Rocks—Two potential LLW repository Taiwan host rocks	Prof. WU, Ming-Chee	
[194] Selected Elements content in Paraguayan Wheat	Prof. FACETTI - MASULLI, J F	
[121] Research Alliance for Validation of PGAA Actinide Nuclear Data	Mr. GENREITH, Christoph	

Thursday 20 September 2012

Session 10 - Radioanalytical Chemistry and Nuclear Analytical Techniques - (08:00-11:30)

- Conveners: Prof. Homonnay, Zoltan (Eotvos Lorand University, Hungary); Prof. Kucera, Jan (Nuclear Physics Institute ASCR, Czech Republic)

time	[id] title	presenter
08:00	[128] OPENING LECTURE - Prompt Gamma Activation Analysis using High-Flux Cold Neutron Beam	Dr. REVAY, Zsolt (Technische Universität München, Germany)
08:20	[228] INVITED LECTURE - Neutron activation analysis: a consolidated analytical tool in the sugarcane agroindustry	Prof. DE NADAI FERNANDES, Elisabete (Nuclear Energy Center for Agriculture, University of Sao Paulo, Brasil)
08:40	[58] INVITED LECTURE - Neutron Activation Analysis and Reference Materials – Development and Perfection	Dr. ZEISLER, Rolf (NIST, USA)
09:00	[153] INVITED LECTURE - Error, Uncertainty, and Metrology in Nuclear Analytical Methods	Dr. LINDSTROM, Richard (National Institute of Standards and Technology, USA)
09:20	[218] INVITED LECTURE - Studies of Trace Element Species in Macromolecules and Protein Nanoclusters by Nuclear and X-Ray Techniques	Prof. CHATT, A. (Dalhousie University, Canada)
09:40	[177] ORAL PRESENTATION - Analysis of Radioactive Waste Waters and Sludges int he Hungarian VVER NPP Paks	Prof. PÁTZAY, György (BME KKFT, Hungary)
09:55	[154] ORAL PRESENTATION - Nuclear Forensics: age determination by the 231Pa/235U ratio	Dr. MENDES, Mickael (CEA, France)
10:10	[173] ORAL PRESENTATION - Prompt Gamma Activation Analysis close to Detection Limits	Dr. KUDEJOVA, Petra (Technische Universitaet Muenchen, Forschungsneutronenquelle Heinz Maier-Leibnitz (FRM II), Garching, Germany)
10:25	[175] ORAL PRESENTATION - Enhancing the dynamic range for high boron concentrations in low neutron capture cross-section matrices with Prompt Gamma Activation Analysis	Mr. SOELLRADL, Stefan (Paul Scherrer Institute & Universität Bern, Switzerland)
10:40	[44] ORAL PRESENTATION - Comparison of Quantitative Neutron Capture Radiography, Inductively Coupled Plasma Mass Spectrometry, and Prompt Gamma Activation Analysis for Boron Determination in Biological Samples	Dr. SCHÜTZ, Christian (Institute for Nuclear Chemistry, University of Mainz, Fritz-Strassmann-Weg 2, D-55099 Mainz, Germany)
10:55	[80] ORAL PRESENTATION - On the development of a rapid method for the concentration and separation of radiostrontium from water samples based on a new Sr selective resin	Dr. HAPPEL, Steffen (TrisKem International, Bruz, France)
11:10	[54] ORAL PRESENTATION - Separation of Uranium and Polonium in drinking water by calix[6]arene columns	Dr. BOUVIER-CAPELY, Céline (IRSN/PRP-HOM/SDI, France)

Coffee break - (11:30-12:00)

Session dedicated to Exhibitors - (12:00-13:00)

- Conveners: Prof. Bonardi, Mauro L. (UNIMI and INFN - Milano, Italy)

Lunch - (13:00-14:00)

Session 11: Education and training in radiochemistry and dissemination of culture in nuclear and radiochemistry (session organized by CINCH consortium) - (14:00-15:35)

- Conveners: Dr. Zeisler, Rolf (NIST, USA); Dr. Cuttone, Giacomo (Istituto Nazionale di Fisica Nucleare, LNS, Italy)

time	[id] title	presenter
14:00	[141] OPENING LECTURE - CINCH - Cooperation in education In Nuclear Chemistry	Prof. JOHN, Jan (Czech Technical University in Prague, Brehova 7, 115 19 Prague, Czech Republic)
14:20	[248] INVITED LECTURE - Nuclear and radiochemistry education in European Universities	Prof. LEHTO, Jukka (Laboratory of Radiochemistry, University of Helsinki, Finland)
14:40	[267] INVITED LECTURE - Nuclear and Radiochemistry Training in the European system of Accumulation and Transfer of Credits for Vocational Education and Training in Europe (ECVET)	Dr. RETEGAN, Teodora (Chalmers University of Technology)
15:00	[265] INVITED LECTURE - Skills and Knowledge Structure Needs - End-users' View	Prof. HANSON, Bruce (University of Leeds)
15:20	[38] ORAL PRESENTATION - Curriculum Development for Nuclear Fuel Chemistry, Reprocessing and Separation Chemistry, and Radioactive Waste Management at the Pennsylvania State University	Prof. UNLU, Kenan (The Pennsylvania State University, USA)

Panel Discussion: Minimum Requirements for a Master's Degree in Nuclear and Radiochemistry - Towards European Master's Degree - (15:35-16:35)

- Conveners: John, Jan (CTU Prague; Czeck Republic)

time	[id] title	presenter
15:35	[266] Panel Discussion - ● Introduction: Prof. J. Lehto; ● Panelists - 3 minutes per panel member; ● Discussion: 10 minutes; ● Panel Summary: 5 minutes	SHI, Weiqun (Chinese Academy of Sciences) REICH, Tobias (Johannes Gutenberg-Universität Mainz) BONARDI, Mauro L. (UNIMI and INFN - Milano) YOKOYAMA, Akihiko (Kanazawa University) NARBUTT, Jerzy (Institute of Nuclear Chemistry and Technology) ALIEV, Ramiz (Moscow State University) TÜRLER, Andreas (Paul Scherrer Institute & Bern University) NITSCHE, Heino (UC Berkeley)

Boat Trip with Gala Dinner - (17:15-23:00)

Friday 21 September 2012

Session 12 - Radioactive elements in the environment, radiation archeometry and Health Physics - (08:00-10:40)

- Conveners: Prof. Ioannidou, Alexandra (ARISTOTLE UNIVERSITY OF THESSALONIKI, Greece); Prof. Groppi, Flavia (LASA, Università degli Studi di Milano and INFN-Milano, Italy)

time	[id] title	presenter
08:00	[60] INVITED LECTURE - The Behavior of Actinide Elements in Contaminated Environments	Prof. CLARK, Sue (Washington State University, USA)
08:20	[216] INVITED LECTURE - Chemical dosimetry for BNCT mixed radiation field and conformal radiotherapy	Prof. GAMBARINI, Grazia (Università degli Studi di Milano, Italy)
08:40	[232] INVITED LECTURE - Determination of the isotopic ratio $^{236}U/^{238}U$ in environmental samples	Prof. WALLNER, Gabriele (Inst. of Inorganic Chemistry, Univ. of Vienna, Währingerstr. 42, A-1090 Vienna, Austria)
09:00	[90] INVITED LECTURE - Study of neptunium sorption on clay and clay minerals using X-ray absorption spectroscopy	Prof. REICH, Tobias (Johannes Gutenberg-Universität Mainz, Germany)
09:20	[229] INVITED LECTURE - Trace Element Analysis with high sensitivity spectrometry.	Dr. PREVITALI, Ezio (Istituto Nazionale di Fisica Nucleare, Sez. Milano Bicocca, Italy)
09:40	[113] ORAL PRESENTATION - Coupling actinide speciation and thermodynamics: Neptunium(VI) solubility and speciation in alkaline NaCl solutions	Dr. ALTMAIER, Marcus (Karlsruhe Institute of Technology, Institute for Nuclear Waste Disposal, Germany)
09:55	[57] ORAL PRESENTATION - Mobilisation of radionuclides and heavy metals from mill tailings at a former uranium mine in south east Finland	Ms. TUOVINEN, Hanna (University of Helsinki, Finland)
10:10	[161] ORAL PRESENTATION - Radioanalytical determinations and radioactivity measurements in the field of Radiation Protection: the experience of the Integrated Laboratory of Radioactivity Measurement and Monitoring of the ENEA Radiation Protection Institute	Dr. ARGINELLI, Dolores (ENEA-Italian National Agency for New Technologies, Energy and Sustainable Economic Development, Radiation Protection Institute, Integrated Laboratory of Radioactivity Measurement and Monitoring, Italy)

Coffee break - (10:40-11:00)

Session 13 (cn't of Session 12) - Radioactive elements in the environment, radiation archeometry and Health Physics - (11:00-13:40)

- Conveners: Prof. Wallner, Gabriele (University of Vienna, Austria); Prof. Clark, Sue (Washington State University, USA)

time	[id] title	presenter
11:00	[73] OPENING LECTURE - Radioanalytical techniques in natural environmental radioactivity determination	Prof. SMODIŠ, Borut (Jožef Stefan Institute)
11:20	[203] INVITED LECTURE - Distribution and transfer of radionuclides including iodine-131 in Japanese environment following the Fukushima nuclear accident	Prof. MURAMATSU, Yasuyuki (Gakushuin University, Japan)

11:40	[108] INVITED LECTURE - Japanese Green Tea: radioactivity measurements, radiochemical extraction yield determination and some radioprotection considerations.	Dr. MANERA, Sergio (LENA - University of Pavia, Italy)
12:00	[65] INVITED LECTURE - Preservation of Cultural Heritage by Radioanalytical Techniques	Prof. NAVARRETE, Manuel (Faculty of Chemistry, National University of Mexico, Mexico)
12:20	[244] INVITED LECTURE - Detecting and evaluating minimal traces of radioisotopes in environment and foods	Prof. RANDACCIO, Paolo (University of Cagliari, Italy)
12:40	[122] ORAL PRESENTATION - Isotopic composition of uranium in aerosol samples collected at 120 km south-southwestern of Fukushima before and after the nuclear power plant accident	Dr. SHINONAGA, Taeko (Helmholtz Zentrum München, German Research Center for Environmental Health, Institute of Radiation Protection, Germany)
12:55	[2] ORAL PRESENTATION - Fukushima fallout at Thessaloniki, Greece (40ºN) and Milano, Italy (45ºN)	Prof. GROPPI, Flavia (LASASegrate, Università degli Studi di Milano, Italy)
13:10	[34] ORAL PRESENTATION - Plutonium radionuclides in stratospheric and tropospheric air: new evidences from measurements in high altitude aerosols	Dr. CORCHO ALVARADO, José Antonio (Institute of Radiation Physics, Lausanne University Hospital, Switzerland)
13:25	[95] ORAL PRESENTATION - Application of microbeam synchrotron techniques to determine the distribution and speciation of plutonium after uptake by Opalinus Clay	Dr. AMAYRI, Samer (Johannes Gutenberg-Universität Mainz, Institute of Nuclear Chemistry, 55099 Mainz, Germany)

Lunch - (13:40-14:30)

Closing Cerimony - (14:30-16:00)

NRC-8, EuCheMS International Conference on Nuclear and Radiochemistry

Sunday 16 September 2012 - Friday 21 September 2012

Como, Italy

Book of abstracts

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Lectio Magistralis / 0

LECTIO MAGISTRALIS - Chemistry in Italy during the late 18th and 19th Centuries

Prof. BELLOBONO, Ignazio Renato

In the first part of the presentation, the birth of ELECTROCHEMISTRY (1799-1800; March 20th, 1800) and the entirely innovative work of **Alessandro Volta**, based on the conversion of chemical into electrochemical energy, by what will be called *pila di Volta* (electrochemical cell), followed by the reverse process studied by **Luigi Valentino Brugnatelli** (electrolysis and electrolytical cell) will be outlined. The epistemological significance of this discovery, which even preceded the atomic theory, and which will be followed, just one century later (December 14th, 1800), by the revolutionary Max Planck's idea of the energy *quantum*, will be discussed, leading us to the second part of the talk.

About 40 years before the birth of RADIOCHEMISTRY, a great debate was taking course in CHEMISTRY, concerning the essence itself of this new-born science, from the question of atomic and molecular weights to periodical properties of the elements and their compounds. Even if about further four decades before, the **Avogrado**'s principle, based on **Gay-Lussac**'s work, had been established, a great confusion existed. The first to have envisaged the potentiality of Avogadro's principle and its correctness has been a young researcher, **Stanislao Cannizzaro**, born in Palermo (July 13th 1826), who won the chair of chemistry in 1855, at the University of Genoa. In this University, he published in 1858 *Sunto di un Corso di Filosofia Chimica*, in which he gave the outmost value to Avogadro's principle, by considering that if two gases, in the same temperature and pressure state, contain the same number of molecules, the ratio of their two volumes gives directly the ratio of their molecular weights (what we now call *formula weights*).

During the presentation of these principles by Cannizzaro, four years after Avogado's death, at the first International Congress of Chemistry of Karlsrule, in 1860, where also **Dmitrij Ivanovič Mendeléev** and **Julius Lothar Meyer** were present, the latter addressed to Cannizzaro a well known appreciation (*we were blind, and you gave us back our sight*). Interpretation of CHEMISTRY on the light of Periodic Table of elements was the necessary presupposition, 36 years later, to the birth of RADIOCHEMISTRY.

In the third part of the talk, some leading Italian chemists, organic chemists particularly, who contributed to the growth of Chemistry throughout the 19th century, will be presented.

At the end, the epistemological connection with the outstanding development of Science in the early decades of the 20th century will be briefly commented.

Opening Ceremony / 263

A warm welcome from the Division of Nuclear and Radiochemistry (DNRC) of EuCheMS

Prof. GAEGGELER, Heinz [1]

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DNRC is one of 18 divisions or working parties that form the European Association of Chemical and Molecular Sciences (EuCheMS), founded in 2004. EuCheMS is an umbrella organisation of European National Chemical Societies promoting the outreach of chemical science and technology to our society as well as to all relevant political organisations. Therefore, the headquarter of EuCheMS is in Brussels.
DNRC represents the field of nuclear sciences in EuCheMS in its manyfold facets, from basic research to a broad field of applications. EuCheMS organizes a congress every second year; the last one was very recently between 26 and 31 August 2012 in Prague (4ECC).
DNRC contributed to 4ECC with three sessions, namely on i) Fukushima & Chernobyl, ii) Applications of Radiotracers to Environmental Sciences; and iii) Speciation of (mostly) Actinide Compounds. These three sessions were part of a Symposium on Chemistry and Environment.
Each division of EuCheMS has the obligation to organize a European conference in its field every second year. DNRC decided to take already existing conference series to meet this requirement, the Nuclear and Radiochemsitry Conferences (NRC) und the Radiochemistry Conference Series (RadChem) organized by our Czech colleagues in Marianske Lazne. Both Conferences Series are organized every four year.
NRC was initiated in 1984 by Prof. K.H. Lieser from Germany. Since then it was hosted by France, England, Austria, Switzerland, Germany and Hungary. We are very happy that our Italian Radiochemistry colleagues invited NRC8 to be held in their home country. We are looking forward to a stimulating and scientifically challenging conference discussing the forefront achievements in nuclear chemistry and in radiochemistry in the beautiful environment of Como.

Session 1 - Radiopharmaceutical Chemistry (radiodiagnostics, radiotherapy, theragnostics) / 250

OPENING LECTURE - Future prospects in diagnostic and therapeutic nuclear medicine

Prof. CHATAL, Jean-François [1]

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For the last two decades a technological revolution has deeply changed the field of application of nuclear medicine. Today PET/CT imaging using 18F-Fluorodeoxyglucose (FDG) is considered as the standard imaging technique in oncology for tumor staging, detection of recurrence and early evaluation of response to treatment. PET/MRI could shortly become the standard imaging technique in neurology. Molecular radiotherapy has also made great progress with the rapid development of radioimmunotherapy (RIT) and radiopeptide therapy (RPT).

For diagnostic imaging, FDG radiopharmaceutical is the only one to be used in routine practice but a lot of fluorinated compounds are currently clinically evaluated and some of them will be approved in the coming years. In oncology several fluorinated radiopharmaceuticals, evaluated in ongoing clinical studies, will allow to visualize tumor functions such as apoptosis, tumor neo-angiogenesis, hypoxia etc… and then to better determine optimal therapeutic strategy. In neurology FDA approved in 2012 Amyvid™ (Florbetapir F 18 Injection) for use in patients being evaluated for Alzheimer's disease and other causes of cognitive decline. Amyvid is thus the first radioactive diagnostic agent approved for PET imaging of beta-amyloid neuritic plaques in the brain.

However the use of fluorine-18 for PET imaging has some limitations and innovative positron-emitting radionuclides are needed. Among them, gallium-68, obtained from a germanium-68/gallium-68 generator and copper-64 with relatively short half-lives and zirconium-89 and iodine-124 with longer half-lives have been the most extentively studied radionuclides and will be routinely used in the next years.

For molecular radiotherapy, efficacy of RIT and RPT has been clearly documented in some clinical settings and new radiopeptides and radioconjugates labeled with lutétium-177, yttrium-90 and perhaps copper-67 will likely be approved for treatment of a panel of cancers especially in the most favorable situation of disseminated microscopic residual disease. Finally the use of alpha particle-emitting radionuclides such as astatine-211, lead-212, bismuth-213 or actinium-225 will be developed for treatment of clusters of malignant cells and isolated tumor cells at the very early stage of cancer progression.

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INVITED LECTURE - A Bridge not too Far: Personalized Medicine with the use of Theragnostic Radiopharmaceuticals

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A major advantage of radionuclides is that they emit radiation of different radiobiological effectiveness and range of action. This offers the possibility of choosing a nuclide the physical and nuclear characteristics of which are matched with a particular tumor type, or the disease under treatment. This presentation introduces a relatively novel paradigm that involves specific individual radionuclides or radionuclide pairs that have emissions that allow pre-therapy low-dose imaging plus higher-dose therapy in the same patient. We have made an attempt to sort out and organize a number of such dual-purpose theragnostic radionuclides and radionuclide pairs that offer this exciting potential of low-dose imaging followed by higher-dose treatment and thus possibly bringing us a major step closer to personalized medicine. This approach would empower the age-long dream of performing individualized or tailored radionuclide therapy in cancer patients, as well as in the treatment of many other disorders that respond to radionuclide therapy. However, an increased and reliable availability of theragnostic radionuclides remains a major issue, which must be addressed before we can successfully put this paradigm into routine clinical practice. A low-dose administration using radiopharmaceuticals based on these theragnostic radionuclides or radionuclide pairs would initially allow molecular imaging (SPECT/CT or PET/CT) to provide the necessary pre-therapy information on biodistribution, dosimetry, the limiting or critical organ or tissue, and the maximum tolerated dose (MTD), etc. If the imaging results then warrant it, it would be safe and appropriate to follow up with dose ranging experiments to allow higher-dose targeted molecular therapy with the greatest effectiveness. It is worth emphasizig that our nuclear medicine modality is the only modality that can fulfill the dream of carrying out tailored personalized medicine by way of enabling diagnosis followed by therapy in the same patient with the same radiopharmaceutical. At BNL, our work on radionuclide therapy has for some time focused on the development of this paradigm for application to several distinct clinical areas, e.g., palliation of bone pain from osseous metastases, treatment of metastatic bone disease, radiation synovectomy, radioimmunotherapy, and cardiovascular applications. This presentation will include the discussion of a number of individual radionuclides and radionuclide pairs which would be potentially excellent choices for theragnostic applications. Since the overall story (including preliminary clinical trials) is a bit more complete in the case of tin-117m, a conversion electron emitter with great theragnostic potential, it will be discussed in more detail, as an example.

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INVITED LECTURE - On the road from Radiopharmacy to Molecular Imaging: the fundamental role of Technetium and Rhenium Chemistry

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Imaging and visualization of molecular events on the cellular and subcellular level requires the design and the syntheses of highly specific compounds which have to to accumulate at desired target sites but should be rapidly cleared from non-targeted organs. Visualization must take place at the living intact subject and the patient.[1] This makes radiolabeled compounds a priority modality for imaging. The design of highly specific compounds does not only concern the biological carrier of the imaging agent as often believed, but the label in particular. Consequently, the label cannot be considered as "just a tag" any longer, but its properties and opportunities have to be integrated in the entire design. Transition metals such as technetium and rhenium (and others) are predestinated to assume roles of structure and function essential molecules due to their inherent diversity of bonds and ligands. Drug finding and development needs, thus, exploration of fundamental chemistry with application in mind. The presentation will outline with examples the importance of fundamental chemistry of technetium and rhenium for applications of concepts and agents. A focus will be put on cyclopentadienyl complexes to emphasize the 3D-space occupation concept. Aspects of the theranostic concept in which technetium complexes are used for imaging and rhenium homologous for therapy will be presented[2] as well as the integration of Tc and Re in very small biological molecules.

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INVITED LECTURE - A renaissance of radionuclide generators for versatile application

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Introduction: Radionuclide generator systems continue to play a key role in providing both diagnostic and therapeutic radionuclides for various applications in nuclear medicine, oncology and interventional cardiology. In parallel to the well established ^{99}Mo/^{99m}Tc generator used for SPECT imaging, new generators for PET/CT imaging attract attention. In particular the ^{68}Ge/^{68}Ga system (^{68}Ge: t½ = 270.95 d; ^{68}Ga: t½ = 67.7 min, β^+ branching = 89.1%) has found impressive and established clinical application, the ^{44}Ti/^{44}Sc generator (^{44}Ti: t½ = 60 a; ^{44}Sc: t½ = 3.97 h, β^+ branching = 94.3) represents a promising system providing a longer-lived daughter, and a ^{140}Nd/^{140}Pr system (^{140}Nd: t½ = 3.37 d d; ^{140}Pr: t½ = 3.39 min, β^+ branching = 51.0%) may be used in terms of an in vivo generator.
Challenges: (1) Different to the established ^{99}Mo/^{99m}Tc generator used for SPECT imaging, new generators for PET/CT such as the ^{68}Ge/^{68}Ga system [1] and the ^{44}Ti/^{44}Sc generator ask for special generator designs because of the long half-life of the parent nuclide. Both represent secular equilibrium systems (different to the transient ^{99}Mo/^{99m}Tc generator) as well as long shelf-life. Both generators may be used over periods of many month and many years, respectively. Still the elution yield of the daughter should be high and the breakthrough of the parent low.
(2) While the generators mentioned all separate parent and daughter nuclides due to different chemical properties of the chemical elements involved, i.e. Mo vs. Tc, Ge vs. Ga, Ti vs. Sc), the ^{140}Nd/^{140}Pr system involves two neighbored lanthanides of very similar chemical behavior.
Experiments and Conclusion: (1) It appears to be a rather general feature of radionuclide generators based on longer-lived parent nuclides, that effective elution modes [2] as well as post-elution processing [3] are mandatory. With those aspects managed in an effective way, the radionuclide generator systems may be tuned to medical systems. This was demonstrated in detail for the ^{44}Ti/^{44}Sc radionuclide generator. Very low breakthrough of ^{44}Ti and high yield of ^{44}Sc is guaranteed by sophisticated generator designs (anion exchange resin as column material, HCl / oxalic acid mixtures for elution), but in addition by a scheme of reverse elution directions. Particular attention was paid to the application of a reverse elution mode, which helps to retain the parent nuclide along the chromatographic column even after many hundred elutions. A 5 mCi generator provides about 170 MBq of ^{44}Sc after direct elution and about 150 MBq following online post-processing similar to the ^{68}Ge/^{68}Ga generator. The final content of ^{44}Ti is as low as < 10 Bq, representing a separation factor of > 10^7..
(2) With no-carrier-added ^{140}Nd produced by irradiations of CeO_2 and Pr_2O_3 targets [4], an efficient ^{140}Nd/^{140}Pr radionuclide generator system was developed and evaluated. The principle of the radiochemical separation is based on physico-chemical transitions (hot-atom effects) of the daughter ^{140}Pr following the electron decay process of ^{140}Nd. The parent radionuclide ^{140}Nd(III) is quantitatively adsorbed on a solid phase matrix in the form of ^{140}Nd-DOTA-conjugated complexes. ^{140}Nd generated is released from the ^{140}Pr-DOTA core as an ionic species. It is easily separated using low volumes of various aqueous eluents. The elution yield is at least 93%, if an optimized eluent, such as DTPA solution is applied. The system remains stable at least over three half-lives of ^{140}Nd, with high radiolytic stability and low ^{140}Nd breakthrough of ca. 0.025% [5].

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INVITED LECTURE - ^{90}Y and ^{177}Lu labelled peptides for PRRT: nuclear and radiochemical aspects

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Somatostatin analogues designed to target tumor cells over-expressing somatostatin receptors have been radiolabelled with 90Y- and 177Lu for peptide receptor radionuclide therapy (PRRT). Clinical trials evidenced large patient variability as regards tumor and organs uptake, thus sustaining the need of tailored dosimetry, for a treatment course with improved safety and efficacy. However, Yttrium-90 does not decay with emission of a γ photon for imaging and cannot be used to estimate radiation dosimetry. Indium-111, which can be used for imaging has been employed as a surrogate. In case of 177Lu-peptide therapy, its gamma-rays enable imaging, dosimetry, and therapy with the same compound.
The radiopharmaceuticals used for PRRT have in common a high renal activity concentration and kidneys have been identified as dose-limiting organs for PRRT, in particular during therapy with 90Y-DOTATOC. Accurate kidney dosimetry plays a key role for the assessment of radiation nephropathy.
The optimal conditions for radiolabelling DOTA-peptides with 90Y and 177Lu have been evaluated. Reaction kinetics were found to be optimal at pH 4-4.5, with a steep decrease at lower pH. The binding kinetics are time- and temperature-dependent, the reactions being completed after 20 min at 80 °C. The highest specific activity (AS) of 90Y and 177Lu correspond to a mol/mol ratio of DOTA over nuclide of 3½ and 6, respectively.
In general, at constant radiolabeling AS the RCP increased proportionally with 177Lu AS, whereas at constant 177Lu AS the RCP steeply decreased with increasing radiolabeling AS.
In conclusion, DOTA conjugated peptides can be efficiently radiolabelled at high AS (>50 MBq/nmol) by 177LuCl3 up to one half-life from production, considering that the standard AS at production of commercially available 177Lu is generally > 740 GBq/mg.

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ORAL PRESENTATION - Radiopharmacological studies of leptin and thyroid hormones relationship in white adipose tissue

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The metabolism of thyroid hormones (TH) in white adipose tissue (WAT) is so far poorly characterized. Therefore, we followed changes in TH metabolism in mice, which were associated with the development of high-fat (HF) diet-induced obesity, with the aid of several radioanalytical methods – radiometric enzyme assays and radioimmunoanalysis (RIA), among others. Possible changes in activities of the key enzymes of TH metabolism in murine WAT were measured not only during obesogenic treatment of the mice but also in response to caloric restriction or leptin administration. Adult male C57BL/6J mice were subjected to these three different treatment protocols. Subcutaneous and epididymal WAT and interscapular brown fat (BAT) depots were dissected and used for morphometric and enzymatic analyses. In addition, blood and samples of liver were also collected (and the latter used as controls for enzymatic determinations). Plasma levels of leptin, as well as total and free thyroxine (T4) and triiodothyronine (T3) concentrations were determined using RIA kits. Enzyme activities of iodothyronine deiodinases of the types 1 (D1), 2 (D2) and 3 (D3) in the liver, WAT and BAT were measured with the aid of our newly developed radiometric enzyme assays. We found that D1 enzyme activity in WAT was stimulated by a high-fat-diet feeding, which also increased plasma levels of leptin. However, D1 or D2 activities in BAT did not change. On the contrary, caloric restriction decreased D1 activity in WAT, but not in the liver, and reduced leptin levels. In return, leptin injections increased D1 activity in WAT. In summary, our results demonstrate, for the first time, changes in D1 activity in WAT under the conditions of changing adiposity, and a stimulatory effect of leptin on D1 activity in WAT. Support from the Academy of Sciences of the Czech Republic (Project No. AV0Z50110509) and from the Czech Science Foundation (GACR Grant No. 304/08/0256) is acknowledged.

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Testing the feasibility of 44mSc/44Sc as a potential in vivo generator for PET imaging and an alternative to the existing 44Ti/44Sc ?

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The use of radionuclides as potential therapeutic radiopharmaceuticals is increasingly investigated. Scandium isotopes (44Sc, 47Sc) become more easily available and their properties are convenient for either PET imaging or radiotherapy. Notably, the half-life of 3.97 h of 44Sc and its high positron branching (94%) lead us to consider this isotope for application in TEP radiopharmaceuticals. The high energy and high intensity ARRONAX cyclotron produces 44Sc together with its isomeric state 44mSc (T1/2=2.44 d) that may stimulate its use as a potential in vivo generator. For in vivo generators, it is not only the log K of complexation between the metal ion and the chelator that is important, but also whether the daughter radionuclide stays inside the chelator after decay of the parent radionuclide. From our previous work [1], we showed that the DOTA chelator exhibit the higher complexation constant value with Sc compared to other often used ligands such as DTPA, NOTA, … In the perspective of medical applications, the Sc(III)-DOTA complex is stable over several days in the presence of a bone mimic and in rat serum. DOTA is also a good chelator of Ho and Nd but against all expectations, it was evidenced that 166Ho, from 166Dy/166Ho in vivo generator was released from DOTA [2]. It was evidenced that was not due to the low recoil energy (Q=0.486 MeV) but due to a "post-effect" as shown previously for the 140Nd/140Pr generator with an even lower recoil energy (Q=0.222 MeV) [3]. The post-effect is attributed to the physico-chemical process occurring after the primary radioactive decay (EC, IT, Auger electron …). This work presents the production route (targetry, extraction and purification) of 44mSc/44Sc at the ARRONAX facility and examines the effect of the recoil energy and the "post-effect" on 44mSc-DOTA complex in the aim of establishing an in vivo 44mSc/44Sc generator. 44mSc/44Sc production, Extraction and Purification For this study, we have produced limited amount of scandium. A typical irradiation corresponds to 30 min at 0.1 µA during 33 MBq of 44Sc are produced. The 44Sc/44mSc activity ratio is found to be 50 at EOB. The extraction/purification process developed lead to a radionucledic purity of 100% and 43Sc, 46Sc, 48Sc<LOD. Synthesis of (radio)metalled complex From 90% to 99 % of radiolabelling yields were obtained for metal-to-ligand ratio ranging from 1/1 to 10/1 respectively; in agreement with published data [1, 4]. Detailed description of data will be given. Study of the Post-effect IT-TOF spectra showed that after a dosis of 1Gy delivered with a 9 MeV electrons beam at the maximum depth dose (external linear accelerator SATURN, ICO-Nantes), the pendant arms of Sc-DOTA complexes opened and the chelate released the metal. A detailed study on the comprehension of mechanisms is will be presented. The experiments with the in-vivo generator are on going. References [1] Huclier-Markai, S, Sabatie, A., Ribet, S., Kubicek, V., Paris, M., Vidaud, C., Hermann, P. and Cutler, C.S. [2011] Radiochim. Acta. 99: 653-662. [2] Zeevaart, J.R., Szücs, Z., Takacs, S., Jarvis, N. and Jansen, D. [2011] Radiochim. Acta 99: 1-5. [3] Zhernosekov, K. P., Folosofov, D.V., Qaim, S.M. and Rösch, F. [2007] Radiochim. Acta 95: 319-327. [4] Majkowska-Pilip, A. and Bilewicz, A. [2011] J. Inorg. Biochem. 105: 313-320

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ORAL PRESENTATION - ^{44,43}Sc and ^{47}Sc as matched pair for theranostic approach to peptide receptor radionuclide therapy

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Recently, great effort is put in development of personalised treatment, including precise diagnoses and therapy using the same molecular targeting vectors. Scandium radioisotopes give opportunity to obtain PET/CT images using 44Sc ($\tau 1/2$ = 3.92 h) and 43Sc ($\tau 1/2$ = 3.89 h) and to treat tumours with a low energy β--particles using 47Sc ($\tau 1/2$ = 3.35 d). The aim of our work was to developed simple production procedures of those radionuclides and to compare 4xSc-DOTATATE labelling efficiency.

Highly enriched 44CaCO3, 47TiO2 and super pure natCaCO3 were used as a target materials. 44Sc was obtained in 44Ca(p,n)44Sc and 43Sc in 40Ca(α,p)43Sc reaction in Joint Research Centre (Ispra, Italy), while 47Sc was produced in research reactor "Maria" (Świerk, Poland) in 47Ti(n,p)47Sc reaction. CaCO3 targets were dissolved in HCl and an ion exchange resin Chelex 100 was used to separate 44/43Sc from target material. The irradiated 47TiO2 was dissolved in HFconc (80 oC). An anion exchange bed, Dowex® 1X8, was used to separate 47Sc from 47Ti. For additional purification of 47Sc, cation exchange column Dowex® 50WX4 was used. 4xSc-DOTATATE was synthesised with different amounts of the peptide and in different pH.

The separation on the Chelex 100 and Dowex® 1X8 resins are efficient. We received high yield of DOTATATE labelling with the three radionuclides. For 15 nmol of the bioconjugate the labelling yield exceed 99%. We checked also the possibility of peptide labelling with 44Sc without target separation using C18 Sep-Pak® column for purification of the 44Sc-DOTATATE.

The three radionuclides can be produced in amount of several GBq. The proposed production procedures are simple and fast. The synthesis and purification procedure can be simplified using C18 Sep-Pak® columns. The 44,43Sc and 47Sc matched pair gives opportunity to further development of peptide receptor radionuclide therapy.

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ORAL PRESENTATION - TiO_2 nanoparticles as vehicles of ^{212}Pb and ^{225}Ac for internal radiotherapy

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There are only a few a-particle emitting radionuclides that have properties suitable for developing therapeutic radiopharmaceuticals. Unfortunately, all available a-emitters have serious disadvantages: 211At forms weak bond with carbon atoms in the biomolecule and in the case of 212Bi, 213Bi and 226Th short half-life often limits the application of these nuclides. However, the short half-life of 212Bi and 213Bi could be effectively lengthened by binding the parent radionuclide 212Pb (t1/2 = 10.6 h) or 225Ac (t1/2 = 10 d) to a biomolecule, thereby effectively extending the use of short half-life 212Bi and 213Bi. In addition, 212Pb/212Bi and 225Ac/213Bi in vivo generator delivers much greater dose per unit of administered activity compared to 212Bi and 213Bi alone.

In our studies we investigated the properties of TiO2 nanoparticles as potential carriers of 212Pb/212Bi, 225Ac/213Bi generators. The TiO2 nanoparticles have special properties like high specific surface and high affinity for certain cations like mentioned Pb2+, Bi3+ and Ac3+, which are useful during the labelling process. Commercially available (P25 Degussa) and synthesised in our laboratory nanoparticles were used in experiments. The nanoparticles were characterized by SEM and NanoSight techniques.

We obtained high yields of labelling (ca. 99%) anatase nanoparticles in case of 212Pb and 225Ac. Afterwards, the stability of labelled nanoparticles was tested in 0.9 % NaCl,

10-3 M EDTA, solutions of biologically active substances (cysteine, glutathione) and human serum. The leakage of 212Pb or 225Ac and their daughter radionuclides was not significant in any of solutions, even when the incubation time was extended to 24 hours. The obtained results show high stability of labelled nanoparticles and allow to begin further experiments, which are based on modification of the surface by silane compounds which enable binding the biomolecules.

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OPENING LECTURE - Organic PET-Radiopharmaceuticals – Aspects of Previous and Current Labelling Techniques

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Diagnostic radiopharmacy has a long-term development since the early investigations of G. v. Hevesy/ F. A. Paneth and the routine availability of radionuclides. After the age of gamma-emitting radiopharmaceuticals, which are still the working horses of nuclear medicine, in the late seventies a new radiochemical/radiopharmaceutical progression started: The era of Positron Emission Tomography (PET) accompanied by the development of PET-scanners and related reconstruction- and processing-algorithms.
Meanwhile PET is an established routine method – starting with few PET-Centres worldwide coming to a broad application in many countries of the world. This growth was accompanied by various influences to basic labelling techniques.
What are the trends in (PET-) radiopharmaceuticals/labelling techniques?
- For routine use automatable, simple one- or two-step synthesis are preferred
- A continuous increase of activity level in routine production takes place
- Development of targetry for improved yields and radiochemical purity
- Aspects of pharmaceutical production rule the daily life for routine PET (GMP)
- Search for new and simple labelling methods
- Search for highly specific radiotracers
- Introduction of positron emitting metallic radionuclides with medium half life
- Combining therapeutic and diagnostic isotope pairs for therapy and dose estimation

These developments are pushed on by applying PET not only for functional diagnostics but also by follow up diagnostics, the need for quantification of physiological parameters and the acceleration of drug application.
In addition, radiochemists are aiming at further evolvement of new labelling techniques based on the progress in – primarily – organic chemistry. Such synthetic pathways have to be simplified, downgraded and speeded up mainly driven by the short half-life of the radionuclides applied. That needs a high level of automation and low substance amounts to be handled.

The presentation will cover these topics from the radiopharmaceutical point of view.

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INVITED LECTURE - Novel ^{18}F-Radiochemistry

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Fluorine-18 is a widely used positron-emitting radionuclide in positron emission tomography (PET) because of its ideal decay characteristics. It decays by positron emission (97%) with a relatively low energy of maximum 0.635 MeV ensuring highest possible resolution. The half-life of 109.7 min permits extended syntheses of 18F-labelled radiopharmaceuticals and study protocols. Fluorine-18 is generated either as nucleophilic [18F]fluoride or as electrophilic [18F]fluorine gas ([18F]F2). The latter is generally obtained in carrier added form. This limits the attainable specific activity of 18F-labelled products generated by the electrophilic method. High specific activity, however, is often critical with PET imaging. Thus, the synthesis of no-carrier-added (n.c.a.) 18F-labelled products is practically limited to nucleophilic methods.
Therefore, novel 18F-radiochemistry focuses on more efficient routes for the introduction of [18F]fluoride into organic molecules. New methods are being developed for its introduction into electron rich aromatic rings, e.g. by using different types of iodonium salts or triarylsulfonium salts. Recently, the synthesis of a sophisticated palladium-based, electrophilic 18F-fluorinating reagent was described started from n.c.a. [18F]fluoride which allows the synthesis of electron rich [18F]fluoroaryl compounds. Concerning the 18F-labelling of molecules in aliphatic position new developments were made based on enzymatic 18F-fluorination, the use of ionic liquids and protic solvents acting as catalysts. The application of "click chemistry" and thiol-reactive labelling agents facilitate peptide and protein labelling with n.c.a. [18F]fluoride.
All the above mentioned methods make use of the conventional formation of a C-18F bond. The easier formation of phosphorous-, boron- or silicon-18F bonds led to a variety of new strategies for 18F-labelling of macromolecules.
In summary, current and advancing radiochemical methods and technologies will be presented which make use of n.c.a. [18F]fluoride in the preparation of 18F-labelled radiotracers for application with PET.

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INVITED LECTURE - Labeling of radiopharmaceuticals with Iodine-124 and their clinical applications

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Iodine isotopes (I-123, I-125 & I-131) have greatly contributed toward expanding the applications of isotopes in medicine; these applications range from ex vivo radioimmuno assays to in vivo SPECT imaging. Iodine-124 (t1/2 = 4.2 d; b+ 23%) was long considered an impurity in iodine-123 production, has lately seen a renaissance in its applications as a PET isotope in clinics due to the ability to produce usable quantities on small hospital-based cyclotrons. This presentation will present an overview of current approaches for production and isolation of iodine-124 and methods for incorporation of iodine-124 onto small molecules, peptides and antibodies. Direct labeling and indirect labeling methods for radioiodination of biological molecules will be discussed in detail. The later part of the talk will exemplify the impact of Iodine-124 based PET radiopharmaceuticals in the clinical setting. Specific examples of pre-clinical and clinical applications including [124I]-NaI for measuring MAPK inhibition and sensitization to radioiodine therapy, [124I]-PU-H71 - a small molecule inhibitor of Hsp90 for measuring drug pharmacokinetics and predicting response to therapy and [124I]-G250-a radioiodinated antibody against CAIX antigen for detecting renal cell carcinoma will be presented.

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INVITED LECTURE - Recoil and conversion electron implications to be taken into account in the design of therapeutic radiopharmaceuticals utilising in vivo generators

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The use of radionuclides as potential therapeutic radiopharmaceuticals is increasingly investigated. An important aspect is the delivery of the radionuclide to the target whereby the radionuclide is not lost from the chelating agent. For in vivo generators it is important whether the daughter radionuclide stays inside the chelator after decay of the parent radionuclide. In our previous work, we showed that the classical recoil effect for β decay only applies to decays with a Q value higher than 0.6 MeV. The loss of the daughter nuclide by a DOTA (1,4,7,10-tetraazacyclododecane-1,4,7,10-tetraacetic acid) chelator was measured for the 166Dy/166Ho generator (Q = 0.486 MeV) and the 90Sr/90Y generator (Q = 0.546 MeV) - transition via the Auger process is absent. It was found that 72% of the daughter (166Ho) was liberated from the DOTA chelator, in contrast to our recoil calculations but corresponding to the ratio of transition of holmium atoms via the Auger process. For the β 90Sr/90Y generator a 1% release from the DOTA chelator was recorded as compared to the estimated 10.2% from the β continuum spectrum of 90Sr. The discrepancy between the experimental and theoretically calculated release can be explained by a correction of the chemical bond energy to 4.4 eV.

Reference; JR Zeevaart, Z Szucs, S Takacs, J van Rooyen, DR Jansen. Recoil and conversion electron implications to be taken into account in the design of therapeutic radiopharmaceuticals utilising in vivo generators. Journal of Labelled Compounds and Radiopharmaceuticals, 2012, 55, 115-119

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INVITED LECTURE - Radiochemistry of Astatine-211: Application to Alpha Particle Targeted Radiotherapeutics

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The heavy halogen 211At, first proposed for use in α-particle targeted radiotherapy more than 30 years ago, continues to be one of the most promising radionuclides for this purpose. Its 7.2-h half life provides some flexibility with regard to the range of molecular carriers with compatible pharmacokinetics including antibody fragments, peptides, affibodies and organic molecules. Its diverse chemistry, possessing both halogen and metallic characteristics has permitted its incorporation into a wide array of targeting vehicles. Most strategies have relied on its chemical similarity to iodine to provide a useful point of departure, with astatodemetallation reactions being a notable example. However, the relatively low carbon-astatine bond strength is challenging, and has lead to the exploration of alternative approaches including those involving higher oxidation states of astatine, complex formation and the labelling of boron clusters. Another important issue from a radiochemistry perspective is the need to compensate for radiolysis-mediated effects including destruction of reactants, reaction with solvent, and alteration in astatine oxidation state, that can occur at the activity levels required for targeted radiotherapy in patients. For protein-based targeting vehicles, specific activity also can be a critical particularly in situations where the average number of receptors per tumor cell is relatively low and heterogeneous dose deposition can be problematic. Finally, if 211At-labeled radiopharmaceuticals are to have a meaningful impact, the ability to produce clinically relevant levels of 211At that can be shipped to remote locations in chemically tractable form must be demonstrated. Hopefully, advances in the radiochemistry of 211At will facilitate the initiation of more clinical trials involving this promising α-particle emitting radionuclide. Our own efforts in that regard currently are directed at developing the methodologies required to initiate clinical evaluation of meta-[211At]astatobenzylguanidine and 211At-labeled trastuzumab in patients with neuroblastoma and breast cancer neoplastic meningitis, respectively.

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ORAL PRESENTATION - Polymersomes as nano-carriers for alpha radionuclide therapy

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Alpha radionuclide therapy has a great potential in the fight against cancer as proven by a large number of pre-clinical and clinical studies [1, 2]. In vivo generators capable of delivering a highly efficient cascade of alpha particles are also steadily gaining importance. 225Ac is at the moment the most important radionuclide that can serve as an in vivo generator, providing four alpha particles with a total energy of 28 MeV. However, the alpha emitting daughter nuclides of 225Ac may deviate from their intended destination due to recoil and cause unwanted damage to healthy tissue. Here, we demonstrate that polymer vesicles (i.e. polymersomes), have great potential to retain the recoiling daughter nuclides based on experimental and simulation data. Experiments reveal that polymersomes with dimensions of 100 nm can easily be loaded with radionuclides and are capable of retaining more than 80 % of the daughter nuclide (209Pb) in the case of 213Po decay [3]. Doubling the size of the nano-carriers increases the retention to 95 %. Furthermore, simulation studies, in which the whole decay chain of 225Ac is considered, indicate that polymersomes can be engineered in such a way that several consecutive recoiling daughters can be retained. According to these results double-layered polymersomes with dimensions of 800 nm will enable the complete retention of the first daughter nuclide 221Fr, while the retention of the third radioactive daughter 213Bi will be increased to 80 %.

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ORAL PRESENTATION - Production of Four Terbium Radioisotopes for Radiopharmaceutical Applications

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Introduction: Terbium provides 4 radioisotopes, which are suitable for different diagnostic or therapeutic applications in nuclear medicine. These are ^{149}Tb ($T_{½}$ 4.1 h, E_{α} 3.97 MeV, I_{α} 16.7 %) for α–radionuclide therapy, ^{152}Tb ($T_{½}$ 17.5 h, $E_{\beta^+ av}$ 1.08 MeV, $I_{\beta+}$ 17 %) for PET, ^{155}Tb ($T_{½}$ 5.3 d, E_{γ} 86.6/105 keV) for SPECT and ^{161}Tb ($T_{½}$ 6.9 d, $E_{\beta^- av}$ 0.154 MeV) for β^--radionuclide therapy. ^{161}Tb is similar to the clinically employed radiolanthanide ^{177}Lu, but emits in addition a number of conversion- and Auger-electrons.

Aim: The production of the four radionuclides should be established in a quality, which is suitable for radiolabeling of biomolecules.

Method: Neutron-deficient ^{149}Tb, ^{152}Tb, and ^{155}Tb were produced at ISOLDE(CERN) by spallation reactions using 1.4 GeV protons onto a 50 g/cm^2 thick tantalum target followed by online mass separation and deposition onto carrier foils.
^{161}Tb was obtained from neutron irradiation of highly enriched ^{160}Gd targets at the high flux reactor of ILL, Grenoble or spallation neutron source SINQ, PSI.
Chemical separation of Tb radioisotopes was accomplished by cation exchange chromatography using the complexing agent α-hydroxyiso-butyric acid (α-HIBA).

Results: 6 MBq ^{149}Tb, 18 MBq ^{152}Tb and 9 MBq ^{155}Tb were obtained in ≤ 1 mL 0.15 M α-hydroxyisobutyrate solution.
Up to 10 GBq ^{161}Tb were produced from irradiated ^{160}Gd targets and obtained in 300 µL 0.05 M HCl.
Radiolabeling of a novel DOTA-folate conjugate was performed with > 96 % yield at Tb:DOTA-folate molar ratios 1:~59000 (^{149}Tb), 1:~5500 (^{152}Tb), 1:~700 (^{155}Tb) and 1:17 (^{161}Tb).
Radiolabeling of DOTATATE was performed with > 99 % yield at ^{161}Tb:DOTA-Tyr^3-octreotate molar ratios ≥ 1:6.

Conclusion: Successful production of the Tb-radioisotopes enabled synthesis of radiolabeled DOTA-folate conjugates and their evaluation in an in vivo study.
Test radiolabeling of DOTATATE confirmed the high specific activity of the obtained ^{161}Tb.

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ORAL PRESENTATION - Decay data measurements on ^{213}Bi using recoil atoms

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^{213}Bi is one of the most important α-emitting; nuclides used in targeted alpha therapy (TAT) against cancer. It is readily available from the subsequent α-decay; of ^{225}Ac → ^{221}Fr → ^{217}At → ^{213}Bi. The parent half-life is $T_{1/2}(^{225}Ac)$=9.920 (3) d (Pommé et al., in press), while ^{221}Fr and ^{217}At are shorter-lived. ^{213}Bi has a half-life of about 45.6 min and decays to the longest-lived alpha emitter ^{209}Bi through two branches, each involving one α-decay; and two β-decays.; An IAEA Coordinated Research Project has identified the need for a new half-life measurement of ^{213}Bi.
In this work, ^{213}Bi has been separated from an open 225Ac source by collecting recoil atoms onto a glass plate in vacuum. The activity of such recoil sources has been followed as a function of time, using an ion-implanted planar Si detector in quasi-2 geometry, resulting in a new half-life value. Additional high-resolution alpha-spectrometry measurements were performed at a solid angle of 0.4% of 4π sr, to verify the energies and emission probabilities of the α-emissions; from the decay products of ^{225}Ac. For both experiments, a description of the measurement method and data analysis is provided. The resulting decay data are given with an uncertainty budget and compared with literature values.

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ORAL PRESENTATION - Bioconjugated nanozeolites labeled with ^{223,224,225}Ra

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Ra-223, Ra-224 and Ra-225, among others alpha-emitting radionuclides, have attractive properties for use in targeted radionuclide therapy. Unfortunately, Ra2+ cations like other cations of the II group, forms very weak complexes, therefore labeling of the biomolecules with 223,224,225Ra is a very difficult task. On the other hand, some zeolites exhibit very high affinity for Ra2+ cations.
In our studies we propose to use nanozeolite as a carrier for transportation of 223,224,225Ra radionuclides to the tumor sites. Due to high ion exchange affinity, nanozeolites strongly bind radium cations and additionally, as aluminosilicates, nanozeolites could be easily attach to biomolecules by silanol-amine/carboxyl linkers.
Nanozeolite of type A (20-70 nm diameter) were synthesized by hydrothermal method. Next, the surface of the nanozeolites was modified by attaching polyethylene glycol (500 - 600 daltons) containing silanol group on the one end and active ester on the second end. The obtained samples were examined for encapsulation of 224Ra and 225Ra (milked from 228Th/224Ra and 229Th/225Ra generators). The stability of radiolabeled nanozeolites were examined in 0.9% NaCl, 0.02 M PBS, 0.001 M cysteine and glutathione and human serum. In the case of 224Ra, about 2% of gamma-radioactivity was found in the solution, which has been attributed to 212Pb - the decay product of 224Ra, while, in the case of 225Ra even above 15% of activity was found in the solution. The activity has been ascribed to 225Ac and 213Bi - the decay products of 225Ra. In the next steps we plan to stabilize 225Ra and all daughter radionuclides in nanozeolite structure. After that, the conjugation of the modified nanozeolites with two vector peptides – octreotide and substance P will be made.

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ORAL PRESENTATION - Development of ^{44}Sc production for radiopharmaceutical applications

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Introduction: 44Sc with a half-life of 3,97 h and a positron branching of 94 % is a promising nuclide for novel PET-radiopharmaceuticals. With an oxidation state of +3 44Sc may be used for radiolabeling of biomolecules with chelators established for coordination of lanthanides such as 177Lu and for other clinically employed radionuclides such as 90Y, 111In or 68Ga. At the Paul Scherrer Institute we are evaluating and optimizing the production of 44Sc for radiopharmaceutical applications.

Methods: 44Sc was produced from a 44CaCO3 solid target at a cyclotron with protons up to 15 MeV (30-50 μA, 20-40 min). For the isolation of 44Sc from the target material extraction chromatography and cation exchange chromatography were employed. After the isolation and purification of 44Sc, radiolabeling reactions were investigated using different molar amounts of a DOTA-folate conjugate.

Results: The irradiation yield was 150 – 200 MBq/μAh with < 1% of the co-produced longer-lived 44mSc (t1/2 = 2.44 d). The chemical separation and purification steps were achieved within 10-15 min providing 80-90 % of the total amount of produced radioactivity of 44Sc in a volume of 0.5 mL, suitable for direct radiolabeling reactions. Radiolabeling of a DOTA-folate resulted in yields up to 90 % .

Conclusions: Rapid column-based chemical isolation of 44Sc from calcium targets and purification provides this novel PET isotope in a form suitable for labeling of biomolecules allowing the performance of in vitro studies. However, to achieve a higher specific activity concentration for in vivo studies further optimization of both, separation and labeling processes, are necessary.

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Electrosynthesis of electrophilic n.c.a. 18F-fluorinating reagents

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Because of its extraordinary suitable decay properties fluorine-18 is the most widely used radionuclide in positron emission tomography (PET) [1]. Presently the radioorganic syntheses of no-carrier-added (n.c.a.) 18F-labelled products are practically limited to nucleophilic procedures. This complicates or excludes n.c.a. syntheses of many putative tracer compounds and intensifies a special demand for electrophilic 18F-labelling. There is the unanswered question whether an electrochemical oxidation of nucleophilic [18F]fluoride can lead to an electrophilic [18F]fluorine analogue. Since in organic fluorochemistry N-F compounds are known as highly effective and selective electrophilic fluorinating agents [2], they were chosen to be synthesized with n.c.a. [18F]fluoride.

Based on cyclic voltammetric measurements an electrochemical synthesis of [18F]N-fluorobis(phenylsulfonyl)amine was attempted. Bis(trifluoromethylsulfonyl)imide with 1-butyl-1-methylpyrrolidinium as counter anion was chosen as a putative suitable precursor. Due to its excellent physical and electrochemical properties this ionic liquid serves as conducting salt, solvent and starting material at once and offers a simplified performance by avoiding major side reactions. Cyclic voltammetry of bis(trifluoromethylsulfonyl)imide showed two succeeding oxidation steps. The first step may lead to a resonance-stabilized radical before further oxidation causes the generation of an unstable cation and finally the decomposition of the compound.

Further electrosynthetic experiments were carried out in an established electrochemical cell [3]. After intercalating [18F]fluoride from aqueous solution into glassy carbon (Sigradur®G) the ionic liquid was added. At a tension of 6-8 V intercalated fluoride and bis(trifluoromethylsulfonyl)imide were expected to co-oxidize and recombine at the surface of the electrode. Products were separated from their ionic contents and analyzed by radio-HPLC. An unambiguous identification could not be achieved due to the lack of a reference compound. A resulting n.c.a. organic product, however, showed a similar HPLC-retention with co-injected bis(phenylsulfonyl)imide as “pseudostandard”. Thus, in principle a production of an n.c.a. electrophilic 18F-fluorinating reagent starting from [18F]fluoride appears probable.

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Gallium-68 complexes of NOTA-bis(phosphonates) conjugates as radiotracers for bone imaging with PET

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This work reports on synthesis, complexation and radiolabeling study of new macrocyclic ligands for selective complexation of gallium, which might serve as potential radiopharmaceuticals for 68Ga-PET bone imaging. Bone-targeting bis(phosphonic) acid moiety, as a distant, non-coordinating group was appended to the 1,4,7-triazacyclonone-1,4-diacetic acid macrocyclic fragment through acetamide or methylphosphinic spacer. Complexation of Ga(III) was studied under different temperature and pH levels by means of 71Ga, 31P and 1H NMR spectroscopy. Complex formation proceeds through intermediate steps involving bis(phosphonate) coordination. Hydrolysis of amide bond of the carboxoamidebis(phosphonate) was also observed during the complexation reaction, leading to the Ga(III)-NOTA complex, confirmed by X-ray diffraction. Under all tested conditions, ligand with methylphosphinate linker showed faster complexation rate than the acetamide. Results from NMR studies (milimolar concentrations) were comparable with gallium-68 radiolabeling study (picomolar concentrations). In vitro sorption study showed effective binding of the complexes to hydroxyapatite, which was used as a model of real bone tissue. Selective bone uptake was confirmed by in vivo PET imaging on laboratory rats.

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Radiosynthesis of 18F-Labeled Diclofenac Hydroxy-Derivative as potential micro-PET imaging tracer

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Methods and Materials:
The diclofenac hydroxy-derivative precursor was prepared via five steps synthesis from 2,6-dichloroaniline.
The radio-synthesis was carried out according to the method described by Wang MW et al with some modifications. It started from the displacement of tosyl group from 1,2-bistosyoloxyethane with 18F-fluoride to afford 18F-fluoroethyltosylate followed by fluoroethylation of diclofenac hydroxy-derivative precursor. Both one-pot and two pot methods were studied. Purification of intermediate and final product was carried out with Sep-pak silica cartridge eluting with diethyl ether and dichloromethane respectively.
The analyses of the labeled intermediate 18F-fluoroethyl tosylate and the final product 10-18F-fluorethyloxy diclofenac and its reference compounds were carried out with TLC. The plates were developed with Dichloromethane/methanol (V:V 95/5) solution. The radioactivity on the plates was detected by Radio-TLC Scanner. No HPLC system was needed for the purification of the product.

Results: 18F- Fluorethyloxy diclofenac was prepared via indirect labeling consisting of fluorination of 1,2-bis(tosyloxy)ethane and fluoroethylation of diclofenac hydroxy-derivative. The radiochemical yield at the end of two steps was about 87%.

Conclusion: In conclusion, an efficient and convenient chemical and radiochemical synthesis of the reference standards and target tracer, 18F labeled small molecule 18F- Fluorethyloxy diclofenac as a new PET imaging tracer, have been well developed. The overall radiosynthesis yield of the tracer was 87%.

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Quantification of iodothyronine deiodinases activities, induced in cultured astrocytes by purinergic agonists, with the use of radiometric enzyme assays

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Described are details of our newly elaborated radiometric methods for extremely sensitive determination of enzyme activities of iodothyronine deiodinases (IDs) of types 1, 2 and 3 in homogenates of cultured mammalian cells. IDs catalyze selective 5'- (outer ring) and 5- (inner ring) mono-deiodinations of iodothyronines and play crucial roles in the bio-transformations of thyroid hormones. The novel radiometric assays for IDs were based on the use of appropriate high-specific-radioactivity 125I-labeled iodothyronines as substrates; TLC separation of radioactive products from the unconsumed substrates; film-less autoradiography of radiochromatograms using storage phosphor screens; and quantification of the separated compounds with a BAS-5000 (Fujifilm Life Science Co.) laser scanner. The applicability of our sophisticated radiometric methods was demonstrated by quantifying the alterations of IDs activities induced in cultured rat astroglial cells by a series of purinergic agonists (like ATP, ADP, AMP and adenosine), retinoic acid, and their combination. The newly developed radiometric assays proved to be very sensitive and rapid and, at the same time, reliable and robust. Support from the Academy of Sciences of the Czech Republic (Project No. AV0Z50110509) and from the Czech Science Foundation (GACR Grant No. 304/08/0256) is acknowledged.

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On the way to the synthesis of the first transactinide carbonyl complex

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Until now gas phase chemical studies of transactinides (TANs) focused on simple, inorganic compounds. The harsh conditions behind the target (plasma and heat) in general prevented direct synthesis of, e.g., organometallic compounds. In the last years, this limitation could be overcome by the combination of a physical recoil separator with chemistry setups. [1]
We report here on initial experiments with this technique that focused on metal-carbonyl complexes. Seaborgium hexacarbonyl has been predicted to be stable. [2] The pi-back bonding, characteristic for the metal-carbon bond in carbonyls, should be stronger than in the complexes formed with the lighter homologues due to the relativistic expansion of the d-orbitals.[2] On the way to experiments with Sg, studies of its lighter homologues were performed. Suitable isotopes of Mo were produced in neutron-induced fission of Cf-249 at the TRIGA reactor Mainz. “Hot” recoil atoms formed volatile complexes upon thermalization in a CO containing atmosphere. The complexes could be rapidly transported in the gas stream to counting or gas chromatography setups. [3] At the UNILAC accelerator at GSI, the gas-filled recoil separator TASCA [4] was used to study the synthesis and chemical properties of tungsten and osmium carbonyl complexes, using conditions directly applicable in a TAN experiment. [3]
This new chemical system promises to give access to a new TAN compound class. Furthermore, it could be used to provide clean of, e.g., Sg isotopes with half-lives of at least a few seconds for nuclear reaction and nuclear structure studies. Elucidation of the experimentally found, but theoretically unexplained isomeric state in Sg-265 [5,6] will be the goal of first experiments.

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Liquid-phase Studies of Seaborgium using the Automated Liquid-liquid Extraction system SISAK

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SISAK is an *automated* liquid-liquid extraction system adopted to one-atom-at-time studies of transactinide elements [1]. It is directly connected to flow-through Liquid-Scintillation (LS) detection cells for unique identification of a given transactinide using alpha-alpha correlations and Pulse-Shape Discrimination (PSD) of beta- and gamma-induced events [2]. The system works behind a physical gas-filled preseparator to significantly reduce unwanted products from the target. This method was developed and demonstrated at Lawrence Berkeley National Laboratory using the Berkeley Gas-filled Separator (BGS) [1,2] and ^{257}Rf produced in a ^{50}Ti on ^{208}Pb reaction. It was the first time such a separator was used for a transactinide chemistry experiments and was very successful. Follow-up experiments [3] where performed which firmly established the SISAK system as suitable for liquid-phase studies of transactinide elements, provided a physical preseparator like the BGS is used to eliminate unwanted products.

Recently, a collaboration between researchers at the Japan Atomic Energy Agency´ (JAEA) Advanced Science Research Centre (ASR) at Tokai and University of Oslo was formed to perform liquid-phase chemistry experiments on seaborgium, element 106. The experiments will be performed at the Nishina Centre for Accelerator Based Science, RIKEN, using the GARIS gas-filled separator. Model and development experiments have been and will continue to be performed at the Tokai Tandem-accelerator and the Oslo Cyclotron Laboratory.

The SISAK system will be set up with an electrochemical cell to enable redox experiments to deduce important and basic chemical information about seaborgium, see NRC8 contribution by A. Toyoshima et al. for further details. For this to work SISAK must be adopted to provide equally rapid transfer as the system used at LBNL (average transport time of 20 s), but with lower flow-rate (0.1-0.2 mL/s compared to 0.4-0.5 mL/s used in Berkeley). The major obstacle to achieve this is the transfer and dissolution of the metal-ions attached to KCl aerosol-particles from the gas-jet transport and into the first liquid phase: When the liquid-flow goes down the volume ratio between liquid and gas gets more and more disadvantageous, leading to significant yield loss in this stage. A new device, based on membrane separation, was constructed which in tests at OCL have shown yields above 80 % even for flow-rates down to 0.1 mL/s, something which has never been achieved with the old centrifuge degasser [4]. Due to its simplicity the new device it is also much faster than the old degasser. Work is in progress to finalize the design and implementation with the SISAK system, results which will be reported at NRC8 together with the overall plan for investigating seaborgium.

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Gas-phase chemistry of carbonyl complexes formed in hot-atom reactions with short-lived isotopes of a Cf-252 fission source

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A new chemical system of metal carbonyl complexes was used for studying the gas-phase chemical behavior of Mo, Tc, and Ru isotopes with a low temperature on-line isothermal gas chromatography apparatus. Carbonyl complexes were synthesized using hot atom gas chemical reactions with carbon monoxide and a 252Cf fission source. On-line isothermal chromatography (IC) experiments, on Teflon and quartz surfaces showed that short-lived Mo isotopes can form carbonyl complexes which are very volatile and interact most likely in physisorption processes. Deduced adsorption enthalpies of Mo carbonyls varied between -37±1kJ/mol and -39±1kJ/mol. Furthermore, the adsorption enthalpies of Tc and Ru carbonyls were determined to be -37±2 kJ/mol and -36±2 kJ/mol, respectively. The speciation of studied carbonyls is still missing. The described data give great hope that this compound class might be well suited for future study of nuclear and chemical properties of the transactinide elements seaborgium, (Sg, Z=106), bohrium (Bh, Z=107), hassium (Hs, Z=108), and possibly also meitnerium (Mt, Z=109).

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Characterization of At- and AtO+ species in simple media by high performance ion exchange chromatography coupled to gamma detector. Application to astatine speciation in human serum.

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Astatine is a rare radioelement belonging to the halogen group. Considering the trace amounts of astatine-211 produced in cyclotrons, its chemistry cannot be directly evaluated by usual spectroscopic tools. Analytical tools, provided that they are coupled with a radioactive detection system, may be an alternative way to study its chemistry.
In the present research work, High Performance Ion Exchange Chromatography (HPIEC) coupled to a gamma detector (γ) was used to evaluate astatine species under reducing and oxidizing conditions. Also, to support the reliability of the experiments, a quantitative analysis using a reactive transport model has been done [1].
Under reducing conditions, results confirm the existence of one species bearing one negative charge in the pH range of 2-7.5. With respect to the other halogens, its behavior indicates the existence of a negative ion, astatide At- [2,3]. The methodology was successfully applied to the speciation of the astatine in human serum. Under fixed experimental conditions (pH 7.4-7.5 and redox potential of 250 mV/ENH) astatine exists mainly as astatide At- and does not interact with the major serum components. Also, the method might be useful for the in vitro stability assessment of 211At-labelled molecules potentially applicable in nuclear medicine. Under oxidizing and acidic pH conditions, a peak corresponding to one species holding one positive charge, in agreement with the literature proposed species [4,5], has been shown. It was defined as AtO+. A change in speciation occurs as the pH changes from 1 to 4. Also, starting with At- at pH 7 under reducing conditions, a change in speciation was observed as the potential increases. In both experiments, this species was eluted in the void volume showing neutral species. A complementary experimental competition method based on liquid/liquid extraction and modeling [6,7] has confirmed the exchange of one proton indicating the probable formation of AtO(OH). The thermodynamic hydrolysis constant obtained by quasi-relativistic quantum calculations [7,8] has also confirmed this species.

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Redox studies of the heaviest elements using an electrolytic column apparatus

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Redox studies of the heaviest elements are fascinating because they provide information on the binding energies of the valence electrons which are expected to be influenced by increasingly strong relativistic effects. All heavy elements with atomic number ≥ 101 are produced in heavy-ion induced nuclear reactions. They are only available as short-lived radioisotopes and only as single atoms. This means that standard electrochemical techniques are not applicable to redox studies of the heaviest elements. Therefore, we have developed a novel technique of electrolytic column chromatography. While passing through one column, single ions undergo electrolytic oxidation or reduction and are chromatographically separated according to their oxidation states.

In this paper, we will present the electrolytic reduction of trivalent mendelevium (Md, element 101) and the oxidation of divalent nobelium (No, element 102) using our newly developed flow electrolytic column apparatus. All experiments were carried out at the JAEA tandem accelerator in Tokai. Redox reactions of Md and No, between the 2+ and 3+ states, were clearly observed. Redox potentials of these elements were successfully determined based on observed amounts of the oxidized or reduced state as a function of the applied potential.

In the preparation of future experiment with seaborgium (Sg, element 106), we recently began a study on the reduction of the lighter group-6 homologs Mo and W using an electrolytic column apparatus. Our recent results of the reduction of Mo and W will be also presented.

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On the fast release of tracer elements from metallic hosts – a step towards vacuum chromatography

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The most common way to chemically investigate transactinides since almost 50 years is by using a gas based transport setup. The "gas-jet" method has proved to be very useful as a fast transport medium, allowing investigations of nuclei with half-lives down to about a second. However, for sub-second nuclei gas-jet type transport has quite poor efficiency due to decay losses. Transport in vacuum is much faster and could therefore be a solution to this problem. Vacuum chromatography [1] is an intenisvely discussed and promissing method that may be applied in the near future to investigate some of the heaviest elements known. In a gas-jet system the evaporation residues (EVRs) are stopped in the carrier gas itself. This is of course not possible in a vacuum system. Therefore it is mandatory to find a new method for stopping the EVRs after their production in nuclear fusion reactions. One possible solution would be to use a very thin metal foil or a stack of metal foils where the EVRs are implanted. However, in order not to loose the speed advantage a vacuum based setup, it is necessary that the subsequent thermal release of the EVR from the metal foil is very fast. We have conducted initial experiments, using the lighter group 13 and 14 homologues of E113 and Fl (E114) to determine suiteable catchermaterials and the best methods to achieve fast release.
The model about the heat of mixing from A.R. Miedema [2] was used to determine the potential catcher-materials Ni, Y, Zr, Nb, Mo, Hf, W, and Re. The experiments took place at the Philips cyclotron at Paul Scherrer Institute in Switzerland. Tracer-doped host materials were produced by the implantation of the fusion products from a Zn, Cd, Hg targets that was irradiated with 80-MeV 4He2+. Otherwise, proton induced reactions were used to produce tracers within the catcher materials. Diffusion constants and activation energies could be determined for the various tracer-host material combinations. Using the principles from Crank [3], Borg, and Dienes [4] it was possible to deduce release rates at various annealing temperatures and for different foil thicknesses. A first on-line experiment was performed at the Oslo Cyclotron Laboratory of the University of Oslo using fission products from proton induced fission of uranium.
Using a model applied for the first time by Bakker [5] it was possible to extrapolate diffusion values from the model experiment and from values from literature to predict corresponding properties for transactinides. For a certain set of experimental parameters, such as catcher material thickness, temperature, and time, it is possible to calculate release rates of superheavy elements from the investigated host materials, assuming that the desorption time of the segregated tracer nuclide is not the time determining factor of the release process.

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Automated rapid α/SF detection system for studying aqueous chemistry of superheavy elements at RIKEN

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The extremely low production yields and short half-lives of superheavy element (SHE) nuclei force us to perform rapid, efficient, and repetitive chemical experiments with single atoms. We plan to start the study of the aqueous chemistry of SHEs such as 261Rf, 262Db, and 265Sg using the RIKEN AVF Cyclotron. In this work, we have developed an automated α/spontaneous fission (SF) detection system that can be coupled to various aqueous chemistry apparatuses. This system consists of a storage column of Ta dishes for holding sample solutions, a round table for sixteen Ta dishes, i.e., a sample collection port, and sixteen detector chambers for the detection of α-particles and/or SF fragments. Beside the round table, we set up the appropriate aqueous chemistry apparatus. The SCARA robot (Yamaha YK500XG) picks up a Ta dish from the storage column through suction and positions it on the round table. The table is rotated to position the dish at the desired port for sample collection, and the solution, typically ~200 μL for each dish, is rapidly dried by using hot helium gas and a halogen heat lamp. Then, the robot transfers the dish to the detector chamber, which is equipped with a Si PIN photodiode (Hamamatsu S3204-09) and a preamplifier (Hamamatsu H4083). The detector chamber is closed promptly and evacuated, and −50 V is applied to the detector from a complex module comprising a power source and a gate generator (Vacuum Products GG-10001). The whole system is controlled by a programmable logic controller (Keyence KV-3000), and each action can be triggered by relay contact signals from a separate controller of the chemistry apparatus. The time required to start the measurement after drying the sample is about 5 s. Each detector has a counting efficiency of 36%. The α-energy resolution is about 50-keV FWHM at 5.486 MeV.

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Extraction of Zr and Hf using TBP and TIOA for the chemistry of element 104 Rf

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Chemical properties of transactinide elements (Z ≥ 104), superheavy elements, are unexpected from the properties of its homologues in the periodic table due to strong relativistic effect on the orbital electrons in the heavy elements. Studies on the transactinide elements have, therefore, attracted many scientists. On the other hand, it is very difficult to perform chemical experiments of these elements because of their short half-lives and low production rates. Rapid chemical experiments on “one-atom-at-a-time” basis are needed. Although there are some pioneering researches conducting chemical separations, studies obtaining chemical constants in equilibrium are a few especially in solution chemistry.

In the present study, the distribution ratios in solvent extraction and distribution coefficients in solid-liquid extraction were determined for Zr and Hf as the homologues of rutherfordium (Rf, Z=104) in the triisooctylamine (TIOA)/HCl and tributylphosphate (TBP)/HCl systems by batch method. From these results, we discussed the chloride complexation and chemical species extracted, and determined the experimental conditions for the extraction of Rf. Based on the comparison of these data with those obtained for Rf, we would clarify the properties in the chloride complex formation of Rf.

Two types of rapid extraction apparatuses are under development to obtain the extraction data of Rf in equilibrium. For solvent extraction, a micro-chemical tip, Teflon capillary tube, and micro mixer device were tested as a mixing section of the solvent extraction apparatus using Zr and Hf radiotracers. As a result, it is found that rapid chemical reactions were observed in using the micro-chemical tip. Solid-liquid extraction apparatus which can perform rapidly solid-liquid extraction by batch method was also developed. By this apparatus, time dependence of the distribution coefficients are expected to be obtained for superheavy elements. Performance of this apparatus was checked by the on-line experiment using the accelerator.

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Development of a liquid scintillation detection system for aqueous chemistry of seaborgium

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As relativistic effects on the valence electrons become large in heavy elements, it is interesting to elucidate chemical properties of the transactinide elements. In these chemical experiments, an automated and rapid chemistry apparatus coupled to an α-particle detection system is required. In the present study, we have developed an on-line liquid scintillation detection system to measure solution samples eluted from the chemistry apparatus for aqueous chemistry of element 106, seaborgium (Sg).

We fabricated a liquid scintillation detector which consists of a photomultiplier tube and a glass cell set in a semispherical reflector. The nuclide 265Sg decays by α-particle emission and the daughter nuclide, 261Rf, decays by α-particle emission or spontaneous fission (SF). Therefore, to detect both α particles and SF fragments is required for the determination of Sg. Energy resolution and detection efficiency in α-particle detection were measured using the nuclides 226Ra and 241Am. Response to the SF events was investigated using the nuclide 252Cf. Procedures in on-line measurements are as follows. An emulsifier scintillator was first introduced into the cell. The aqueous solution sample from the chemistry apparatus was then injected to the cell. During the injection, the scintillator and the solution sample were mixed with an electromagnetic stirrer. Measurement was started after the mixture became homogeneous in the cell. The above procedures were automatically and repeatedly performed.

The present detection system was applied to the on-line measurement of 213Fr (T1/2 = 34.6 s) produced in the 209Bi(16O, 4n)221Pa reaction using the AVF cyclotron at RCNP. The reaction products were transported to the chemistry laboratory by a He/KCl gas-jet system and were collected on the collection site of the ion-exchange apparatus, ARCA, for 1 min. The collected products were then dissolved in 200 μL of 0.1 M HNO3/5 × 10-3 M HF solution for 12 s. The eluent was transferred to the cell and mixed with 4 mL of the scintillator. Start of measurement of the products was approximately 15 s after the end of collection.

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Diamond Detectors in Transactinide Chemistry

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The possibilities of on-line thermochromatography experiments with transactinide elements depend on the material of the applied detectors. Si PIN-diodes or Si PIPS-detectors are only usable in a temperature interval between 90 K and 315 K limited by the electronic band gap of Si of about 1.1 eV. In recent experiments using the COLD [1] or COMPACT [2] detector setup, the upper bound of the temperature gradient was limited to 310 K for this reason. Therefore, the experimental characterization of less volatile elements with higher negative adsorption enthalpies is not feasible. Already the rather volatile mercury with a negative adsorption enthalpy on gold of 98 kJ/mol [3] - corresponding to a deposition temperature of about 430 K using the experimental condition of the COLD system - reveals the constrained range of the currently used setups.
In recent years the development of chemical vapor deposition (CVD) single crystal diamonds made a considerable progress concerning the available size, quality, and quantity. CVD single crystal diamond detectors can be heated up to temperatures higher than 500 K without any spectroscopic degradation, due to their electronic band gap of 5.5 eV [4]. In addition, they can be operated under broad daylight or strong IR radiation (e.g. near a heat source) and last but not least, diamond has one of the highest thermal conductivities known, which is important for reproducible properties of the chemical surface temperature.
We prepared, characterized, and tested different types of CVD diamond detector setups, aiming for an application in future transactinide chemistry experiments.

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Extraction behavior of Mo(VI), Mo(V), W(VI), and W(V) from HCl solutions by Aliquat 336

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It is expected that the element 106 seaborgium (Sg) is redox active, because molybdenum (Mo) and tungsten (W), the homologues of Sg, have various oxidation states. Electrochemistry of superheavy elements (SHEs), including Sg, with atomic numbers ≥ 104 is very interesting, because valence electron structure of SHE would be influenced by strong relativistic effect. However, the voltammetry for measurement of redox potential cannot be applied to SHE chemistry, because the SHE nuclides can only be produced as one atom at a time and have short half-lives. Therefore, the investigation of reduction potential of Sg ion should be combined with chemical separation to obtain reduction ratio of Sg. We are planning to study the reduction behavior of Sg by the electrochemical method combined with solvent extraction.

We carried out solvent extraction of Mo(VI), Mo(V), W(VI), and W(V) in 0.01–0.36 M Aliquat 336 / 0.1–11 M HCl system as model experiments for Sg. The HCl solutions of $Na_2[MoO_4]\cdot 2H_2O$, $[MoCl_5]$, and $Na_2[WO_4]\cdot 2H_2O$ were used for extraction of Mo(VI), Mo(V), and W(VI), respectively. The HCl solutions for Mo(V) and W(V) were prepared by electrochemical method and $SnCl_2\cdot 2H_2O$ as reductant, respectively. Extraction behaviors of mononuclear Mo and W were investigated using carrier-free radiotracers ^{99}Mo and ^{181}W, which were produced as ^{235}U(n, f) and ^{181}Ta (p, n) reaction, respectively.

The distribution ratios (D) of Mo(V) and W(V) were higher than those of Mo(VI) and W(VI), respectively. The D values for Mo(V) obtained by the reduction of Mo(VI) were in good agreement with those obtained with $[MoCl_5]$, suggesting that reduction behavior of the group-6 elements can be observed by solvent extraction. The D values of carrier-free Mo(VI) and W(VI) are almost the same as those with macro amounts in 6–11 M HCl. This condition would be suitable for the Sg experiments.

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Study on Quadrivalent Chemical Species of Rutherfordium in Aqueous Solution by Means of TTA resin

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Rutherfordium (Rf) has attracted a lot of attention in research on the chemical properties of a superheavy element. For the purpose of its speciation in aqueous solution, we aim to observe the chemical behavior of Rf by means of reversed-phase chromatography with a chelate extractant of 2-thenoyltrifluoroacetone (TTA) as the stationary phase. It extracts quadrivalent metal ions preferentially, and, that is, it will make possible determination of a specific complex formation constant of Rf.

Prior to the experiments with Rf, batch experiments of Zr and Hf were performed to determine the time to attain the chemical equilibrium on TTA-resin and the eventual distribution ratios. The resin was brought into contact with ^{88}Zr and ^{175}Hf carrier-free atoms in the acid solutions of 9.5×10^{-5} – 0.10 M HF / 0.1 M HNO_3 and allowed to attain equilibration in a polypropylene tube at room temperature. An aliquot of the aqueous phase was subjected to γ-ray spectrometry using a Ge detector. Similarly, an off-line reversed-phase extraction chromatography of ^{88}Zr and ^{175}Hf was also performed in a teflon tube column (1.6 mmφ) or a micro-column (1.6 mmφ×7 mm) into which the resin was filled. Then an on-line reversed-phase extraction chromatography was performed with ^{89m}Zr and ^{175}Hf, simultaneously produced in the ^{89}Y (p, n) and ^{175}Lu (p, n) reactions, respectively, at the RIKEN K70 AVF Cyclotron.

Finally, the chemical system for the Rf experiment with ARCA (automated rapid chemistry apparatus) with the micro-columns was connected to the automated rapid α/SF detection system at the AVF cyclotron and subjected to the experiment with ^{85}Zr ^{169}Hf, and ^{261}Rf isotopes produced in the ^{18}O-induced reaction with the targets of natGe, natGa, and ^{248}Cm, respectively. Thus far we have succeeded in obtaining a preliminary α-spectrum of Rf nuclides transferred through a gas-jet system and passing through the ARCA system. We are now ready to proceed to the further experiments for the purpose of obtaining a better statistical data.

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Characteristics of Uranium species when U(III) in a LiCl-KCl molten salt was leached out with water and ionic liquid

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As a type of spent nuclear fuel treatment, the pyrochemical process is well known for its non-proliferation of nuclear fuel cycles, separation of long-term radioactive nuclides during processing, the recovery of uranium for re-use as a nuclear fuel, and a significant volume-reduction of high-level wastes. After the complete pyrochemical processing is finished, a remaining small amount of salt waste, apart from the salt for recycling purposes, will be stored for the long term and is composed of some actinides and lanthanide species (mainly exist as 3+ ions) dissolved in molten salt. In this study, we investigated the behavior of U(III) dissolved in LiCl-KCl molten salt, when U(III) was leached out with water compared to ionic liquid, to obtain better understandable information for long-term waste salt storage.

A U(III) in LiCl-KCl eutectic salt was prepared from the reaction of uranium metal with cadmium chloride in a LiCl-KCl mixture (44 wt.% LiCl) at 450 oC in an Ar-atmosphere glove box. For a characteristic study of U(III) under stable and unstable conditions, the U(III) in the LiCl-KCl eutectic salt was first dissolved in an appropriate ionic liquid and water.

U(III) is unstable (oxidized) in alkali fluoride molten salt or under general conditions but is stable in LiCl-KCl molten salt. Moreover, the ionic liquid (1-hexyl-3-methyl-imidazolium chloride) used in this research did not cause an oxidation or reduction of U(III) like water does. The behavior of U(III) in LiCl-KCl is dependent on the contacted solvent (ionic liquid or water), and the results, including the actual assignment of each peak of the spectroscopic spectra, were conclusive.

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Selective ion exchangers for Fukushima waste effluent purification

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Two inorganic ion exchangers manufactured by Fortum company, Finland, and developed in collaboration with the Laboratory of Radiochemistry, University of Helsinki, have been selected for the radionuclide removal in a new purification system of Energy Solutions, USA, to be commissioned at the Fukushima power plant in 2012. The system will purify approximately 200,000 m3 of highly contaminated effluents obtained due to pumping of sea water into damaged reactors after tsunami had destroyed controlled water circulation in the reactors. The two ion exchangers are CsTreat® for radioactive cesium removal and SrTreat® for radioactive strontium removal. The former is based on a transition metal hexacyanoferrate and the latter on a sodium titanate. These unique ion exchange materials are produced as granules suitable for column use.

CsTreat® and SrTreat® have been utilised in several industrial radionuclide removal processes during the last twenty years [1]. CsTreat® has been in use at Loviisa NPP since 1991 for the decontamination of high-salt (ca. 200 g/L NaNO3/KNO3) evaporator concentrates and about 1,600,000 litres have been purified with only 230 liters of CsTreat® material with a decontamination factor over thousand [2]. Most recent application of CsTreat® took place in the decommissioning of Dounreay Fast Reactors in the UK [3].

The selectivities of CsTreat® and SrTreat® are much higher than that of common organic resins or other radionuclide-selective inorganic materials. For example, the selectivity coefficient of CsTreat® over sodium (kCs/Na) is 1,500,000, which is four orders of magnitude higher than for zeolites and two orders of magnitude higher than for a silicotitanates [4], both of which have been used in the Fukushima waste effluent treatment. As processing capacity for trace level radionuclide removal is mostly governed by the selectivity, CsTreat® can offer superior performance compared to other Cs-selective materials.

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Poster Session / 21

Experimental Investigation on Cryogenic Adsorption of Low-concentration Hydrogen from Helium by MS5A

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Tritium extraction system (TES) is one of the most important components in the helium cooled solid breeder(HCSB) test blanket modules (TBMs) of ITER. TES will extract various isotopic species of hydrogen by the liquid nitrogen cooled molecular sieve adsorber beds (CMSB). The globular molecular sieve 5A(MS5A) with the diameter of 3~5 mm was selected as the candidate for the CMSB in TBM TES. The MS5A was studied by means of the thermogravimetic analysis(TGA) and thermal desorption spectroscopy(TDS). The adsorption property of the adsorbent was also investigated in continuous He-H2 purge gases with a flow rate of 33.6 L/min. The durableness of MS5A was assessed according to the runtime of ITER. The results show that MS5A can extract traces of hydrogen from helium effectively, furthermore, the content of hydrogen was quite high in the regenerated gases of CMSB after saturated adsorption. The adsorbent can stand the test during the running of ITER. Therefore the MS5A could recover low concentration hydrogen effectively and reliably from the purge gases of the TBM during the operation of ITER.

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The Effect of Cellulose Degradation Products on the Migration of 90Sr in Cementitious Backfill using Radial Diffusion.

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Many concepts for the geological disposal of intermediate level (ILW) and low level radioactive waste (LLW) include backfill materials based on admixtures of Ordinary Portland Cement (OPC). It is expected that these backfill materials will generate high pH conditions and further, the eventual corrosion of the metal canisters used for disposal will promote a low Eh environment. It has been generally assumed that the safety functions of the cement within the near field of a Geological Disposal Facility (GDF) will include reduction of the solubility of many radionuclides and retardation of migration by sorption and incorporation.

Diffusion will remain the dominant migration mechanism for radioisotopes throughout the post closure period. The radioisotope 90Sr will be a significant component of the disposed waste and its half-life of 28.8 years means that escape and migration from the GDF could be of significance if it occurs within the operational phase or first few hundred years, post closure. Cellulosic materials will also be disposed and previous studies have shown that cellulose degradation products (CDP) produced at high pH can enhance the migration of metal ions.

The radial diffusion experimental technique uses small pre-cast cylinders of the matrix under investigation. An appropriate concentration of 90Sr is introduced into a cavity in the centre of the cylinder, which is then sealed and placed in a solution previously equilibrated with the matrix. The increase in concentration of the isotope in the external solution is then determined at defined time intervals.

Diffusion experiments on NRVB (Nirex Reference Vault Backfill) have been undertaken using 90Sr in the presence and absence of CDP or gluconate (a surrogate for the CDP mixture). The effect on the migration of 90Sr caused by addition of CDP is seen to be significant and work is currently underway to understand the mechanisms responsible.

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Recoil-radiolabelling of nanoparticles with 7Be generated by 7Li(p,n)7Be reaction in mixed powder targets.

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Nano-sized materials have highly interesting physico-chemical properties that may be employed to great advantage in many areas. However, their application in various fields of society (e.g. in consumer products, including cosmetics or food) also implies safety issues. Many studies on nanoparticle toxicity have been performed, but a basic problem regarding risk assessment is nanoparticle quantification. This may easily be addressed by using radiotracers and radiolabelled nanomaterials. Some industrially produced nanomaterials (e.g. SiO2, Al2O3, or carbon-based nanoparticles) are
difficult to radiolabel without significant chemical surface modification.

We present here a solution to this difficulty based on recoil light-ion implantation (e.g. Be-7). An overview of different radiolabelling methods as well as the Be-7 ion implantation method is presented. Radiolabelling yields and the influence of irradiation on nanomaterials is also discussed.

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Study of the Production of Mo and Tc Medical Radioisotopes Via Proton Induced Nuclear Reaction on natMo.

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99mTc radioisotope is a very important medical radioisotope for diagnostic tests. In this work an alternative root of producing this isotope, either directly or through the generator 99Mo (99mTc) , namely using cyclotrons, is introduced and discussed. The excitation functions for the different proton-induced nuclear reactions on natMo target are measured and compared with some previously measured data. This study aims to release some contradictions between the existing data, and to give a reliable data set for the production of 99mTc and some other isotopes of importance in nuclear medicine beside some impurities. Some monitoring reactions on Al And Cu targets are also measured and compared with the recommended IAEA data sets, in order to give high degree of consistency to our results. The present excitation functions confirms some previously measured sets, while contradicts with others. Therefore, further expermintal studies are needed to obtain a recommended cross-section values for the production of 99Mo, 94gTc, 95gTc and 99mTc via proton induced reactions. Theoretical code calculations using TALYS code are performed and show a good consistency with the measured cross section values. The code calculations can be used for cross section estimations, when no enough experimental data are existing. Furthermore, the integral or thick target yields are estimated based on the measured excitation functions for all the investigated reactions.

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Synthesis and Characterization of Radiolabelled Silver Nanoparticles

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Silver nanoparticles (AgNPs) have a wide range of applications in cosmetics, wound dressings, food packaging and also in medical sciences such as drug and gene delivery systems because of their effective antibacterial properties [1,2]. This wide usage increases environmental and human exposure to AgNPs which may cause undesirable biological and ecological effects. Recent studies showed that AgNPs have significant toxic effects on cells and the particle properties such as size, surface area, solubility etc. may alter the toxicity of the particles [2-4]. Therefore it is essential to investigate their behaviour in complex matrices. The chemical behaviour of silver and silver ions is in biological and environmental media is extremely complex due to
a)relative easy which silver may be oxidised or reduced
b)photochemical activity of both the metal and its salts
c)the large number of low solubility complexes or compound which may form with silver ions.
All of these factors can create many difficulties and uncertainties when attempting to achieve accurate and reproducible analysis of low levels of silver by conventional trace analysis methods such as ICP-MS. Many of these problems can be reduced or eliminated if silver detection, tracing and quantification can be done using radiotracers. Use of radioactive NPs as tracers has several advantages with respect to the high sensitivity and accuracy of radiolabelling techniques [5].
In this study radiolabelled AgNPs were synthesized by reduction of silver nitrate with sodium borohydride. The 105Ag and 110mAg were prepared by proton and deuteron irradiation of silver foils respectively at the JRC Scanditronix MC 40 Cyclotron, followed by dissolution in nitric acid and evaporation to dryness. Due to a higher production yield, 105Ag was used for radiolabeling studies. The characterization of the nanoparticles was performed using DLS, zeta potential and XRD analysis. The size distribution of the 105AgNPs was found to be centred around 20 nm according to the DLS measurements and were colloidally stable for at least 21 days. It was observed that size distribution and surface charge of the radiolabeled nanoparticles was very similar to those obtained by synthesis using a non-radioactive source of silver. Silver ion leaching and particles stability in different media were assessed.
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Poster Session / 1

Nuclear and radioanalytical techniques in nanotoxicology research: studies on the Rabbit Reproductive System

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Nanotechnology, perceived as one of the key technologies of this century. Despite the term "nanotechnology" is synonymous with things that are innovative and highly promising, little attention has been dedicated to the other side of the coin, i.e. the research on toxicological effects and on the relations with factors that can affect the nanotoxicity on human health and on the environment. Research on the impact of engineered nanoparticles (NPs) is strongly hampered by a lack of reliable tools to detect, visualize and quantitatively trace particles movement and transfer in complex environmental and biological systems. A few methods, such as labelling with fluorescent probes, may overcome some of the detection problems; however, leading a significant modification of the particles to be traced, the engineered NPs behavior is modified. Conversely radiolabelling or radioactivate the NPs to make them distinctive and thus easily detectable, is one very smart way to solve the problem especially because after neutron or proton activation the physicochemical characteristics in terms of size distribution and Z potential are maintained as the “cold” ones. One of the debated themes is the influence of NPs on the human reproductive system. In this contest a way to have experimental evidence is the study of AuNPs and AgNPs passage through the blood-testicular barrier in vivo. In this work some preliminary results obtained by NAA of the sperma (seminal liquid plus sperms) from selected strain of rabbits are presented. Samples were taken from animals exposed for different time (3 and 7 days) to 5 mg AgNPs and AuNPs kg^{-1} b.w.

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Synthesis of new 18F-labelled Porphyrins and their potential aplication for in vivo Molecular Imaging with PET

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Molecular imaging holds the promise of non-invasive assessment for biological and biochemical processes in living subjects using specific imaging tracers. Positron Emission Tomography (PET) is a highly specific and sensitive molecular imaging technique with widespread use for research and clinical application. The majority of PET studies today are performed with molecules labelled with fluorine-18, a radionuclide possessing important characteristics including a favourable half-life (110 min) and the ability to replace H in organic molecules.1

It's widely recognised that porphyrins are one of the most important prosthetic groups in biological systems and porphyrin derivatives have recently found promising biomedical applications in detection and treatment of a variety of tumours due to their affinities for these tissues in relation with the nature of the side chain and the mechanism of their physico-chemical action.2-5 In this communication we describe our recent studies on the synthesis of novel sulfonamide substituted meso-tetraphenylporphyrins and automated synthesis of new 18F-labelled porphyrin derivatives, by fluorination via nucleophilic substitution with K2CO3/K2.2.2/ACN. Preliminary biodistribution studies in rats with PET will be presented. We believe our results may open new directions for the development of new theragnostic tools.

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New analytical method for actinide (Pu, Am, U, Th, Np) separation based on diglycolamide resin (DGA)

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One of the most critical part in the treatment of liquid radioactive wastes is the separation process of tervalent actinides, especially that of Am in acidic solutions. After decades of investigation a new extraction chromatographic material, the DGA resin was developed with good extraction properties for all actinides, moreover extremely high extractability towards Am1.
Our goal was to separate major and minor actinides, i.e. U, Th, Pu, Np, Am (Cm) from radioactive waste samples and determine the radionuclides by α spectrometry. DGA is a good candidate material for the extraction chromatographic separation of actinides in tetra-, hexa- and (moreover) in tervalent oxidation states due to the high distribution ratios from a wide range of concentration of nitric and hydrochloric acid solutions. It shows potential for the simultaneous separation of actinides with the use of a single column.
The distribution ratios of Th(IV), U(VI), Pu(IV) and Am(III) on DGA have been determined in hydrochloric and nitric acid media1. In our laboratory extraction studies were carried out on various oxidation states of the actinides including Np species and on the effect of several possible interferences (Ca^{2+}, Fe^{2+}, Fe^{3+}, SO_4^{2-}, $S_2O_8^{2-}$, $S_2O_5^{2-}$, SO_3^{2-}, N_2H_4, NO_2^-, I^-) by batch and/or column uptake experiments using N,N,N',N'-tetra-n-octyldiglycolamide (DGA Resin, Normal) produced by Triskem ©2. Based on the measured distribution ratios two new methods were developed for Th, U, Pu, Am and Np separation in model solutions. Oxidation state adjustment of actinides was performed before and during on-column separation. The methods have been tested by the analysis of liquid radioactive waste samples originating from nuclear power plant using alpha spectrometry. Performance parameters of the procedures such as chemical recoveries and decontamination factors will be presented.

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Sequential separation and determination of Pu, Sr-90 and Am-241 in soil and sediment samples using DGA Resin for the preconcentration of the actinides.

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A method for the sequential, and quantitative, separation of Pu, Sr-90 and Am-241 radionuclides in environmental soil and sediment samples is presented (oder: has been developed). After wet- and dry-ashing of the samples, Pu and Am-241 were preconcentrated from the leaching solution on DGA resin, whereas Sr-90 was not retained and collected in the eluate. Pu was then separated from Am-241 using an anion exchange resin (BioRad AG 1-X2) and was further purified by UTEVA and DGA resins in series. Am-241 was separated from the lanthanides using a TEVA column. In parallel, Sr-90 was purified by a Ca-oxalate precipitation followed by separation on Sr resin. The measurements of Pu and Am-241 were performed by α-spectrometry. Sr-90 was measured by low-level liquid scintillation counting. The developed method was validated by analyzing IAEA reference materials and environmental soil samples.

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Neptunium Redox Chemistry in Irradiated HNO3 Solutions

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The reliable separation methods for neptunium assume the ability to maintain a preferred oxidation state. However, regardless of its initial redox speciation, a series of reactions occurs in nitric acid to create a mixture of oxidation states including Np(V), Np(VI) and sometimes Np(IV). Additionally, the irradiated solutions such as dissolved nuclear fuel contain both transient and long-lived radiolysis products which may be strongly oxidizing or reducing. Thus, irradiation may be expected to impact the chemical equilibrium and distributions of neptunium of various oxidation states.

Among the transient products of the irradiation of aqueous nitric acid are the reducing •H atom and solvated electron and the oxidizing •OH radical from water radiolysis, and the oxidizing •NO3 and •NO2 radicals from nitric acid radiolysis. Longer-lived radiolysis products include hydrogen peroxide from water radiolysis and nitrous acid from nitric acid radiolysis. While it has been known that HNO2 acts as a reducing agent toward Np(VI); the effects of the oxidizing transient species have been much less studied.

Solutions of neptunium in nitric acid irradiated with Co-60 gamma-rays were analyzed by UV/Vis spectroscopy on radiolytically-induced changes in neptunium valences, as well as the nitrous acid concentration. It was found that at low absorbed doses, the oxidizing radicals oxidized Np(V) to Np(VI). However, as the irradiation proceeded the concentration of nitrous acid became sufficient to reduce Np(VI) to Np(V), and then continued irradiation favored this reduction until an equilibrium was achieved in balance with the oxidation of Np(V) by nitric acid itself. The starting concentrations of the two neptunium valences did not affect the final equilibrium concentrations of Np(V) and Np(VI).

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Determination of Boron distribution in Co-Re alloys

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Co-Re based alloys are being developed at the TU Braunschweig to supplement Ni-based Superalloys at ultra-high temperature (>1200°C) applications. Grain boundaries in these polycrystalline alloys are strengthened by boron. Boron is known to segregate to grain boundaries in Ni-alloys and improve low temperature ductility. The mechanisms to strengthen the grain boundaries are being explored for the Co-Re alloys. To have a better understanding of the effect, a set of experimental alloy was manufactured with known added boron amounts ranging from 50 to 1000 ppm. However, as boron is volatile, the quantity remained in the alloy is presumably lower than added.

The aims of the present experiments were to quantify the boron content by PGAA, and to map its distribution in the alloys, looking for signs of segregation. Thanks to the high cross-section of the 10B(n,alpha gamma)7Li reaction, we could detect boron already in a few ppm quantity, based on its 477.6 keV gamma-ray. A complementary technique, solid state nuclear track detectors (SSNTD) was used to map the near-surface boron density. The alpha particles, emitted from the same nuclear reaction, create tracks in the SSNTD, if we make a close contact between a polished surface of the sample and the track detector during the irradiation. The track detectors were etched in hot NaOH and imaged with an optical microscope. An attempt was also made to have a closer look to the boron-spots with SEM, giving far better spatial resolution than a conventional microscope.

Several measurements have already been carried out at the PGAA beamline of the Budapest Neutron Center. In some samples the segregation was clearly detectable. This information allows the material scientists to develop a new generation of materials.

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Characterization of silicon for photovoltaic applications with INAA and PGAA

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The required impurity level for silicon used in solar cell production can be much higher compared to the demand in the electronic industry [1]. Therefore processes are investigated to clean the raw silicon to an impurity level necessary for photovoltaic applications. Of special interest is the impurity level of the 3d transition metals and of the dopant element boron. Transition metals of the 3d – series form deep levels in the energy gap of silicon and act as recombination centres for the charge carriers thus degrading the efficiency of solar cells.

A major part of the purification of silicon takes place during directional crystallization because most impurities have a segregation coefficient k < 1 [2]. In order to find optimal crystallization parameters, the impurity profile along the crystallization front is of special interest. After cutting the ingots of multi-crystalline silicon into wafer further cleaning procedures like HCl gettering [3] are applied. In this procedure, a wafer is heated to temperatures above 600 °C at which a continuously streaming of inert gas (such as N2) or a reducing gas (H2) containing a few percent of HCl interacts with the wafer.

In this project, instrumental neutron activation analysis (INAA) is used to determine the 3d impurity concentration at the different purification steps beginning with metallurgical grade silicon (MG-Si) which is used as feedstock material for the crystallization and ending with the analysis of silicon wafers after the gettering procedures. Special attention is given to the impurity profile after crystallization. The neutron irradiation is carried out at the research reactor TRIGA Mainz [4] at a neutron flux from 0.7 x 10^12 /cm^2s up to 4 x 10^12 /cm^2s depending on the impurity level of the investigated material. The boron concentration was determined by prompt gamma activation analysis (PGAA) at the HFR research reactor in Petten.

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INAA for discriminationg geographic origin of Brazilian rice

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The concentration of chemical elements in plants and in their edible parts varies according to the plant genotype, soil fertility, environmental factors and agricultural practices. Based on such relation, chemical elements have been used to discriminate the origin of food, identifying species or variety, cultivation system and geographic region, amongst other characteristics. Trace elements are especially interesting for discriminating the geographic origin, since their availability for plants is normally connected to the geology and genesis of soils. As a staple food worldwide, rice has already been studied by different analytical techniques for tracing origin. Here, instrumental neutron activation analysis was applied for evaluating Brazilian rice samples, in order to identify chemical elements with potential for discriminating geographic origin. Sampling was performed directly in the consumer market of Piracicaba city, São Paulo State, tracking the origin back to the processing unit. Emphasis was given to commercial brands from Rio Grande do Sul, the state with the main production of rice in Brazil. At total, twelve municipalities of Rio Grande do Sul were represented by the sampling procedure, comprising the regions west, south and east of the State. After processing in a rotor mill, samples were irradiated at a thermal neutron flux of 8x1012 cm-2 s-1 for 8 hours and radioactivity was counted in HPGe detectors at decay times of 4, 7, 15 and 30 days. Concentrations of chemical elements were assessed by the k0 method, allowing the determination of As, Br, Co, Cs, K, Mo, Na, Rb and Zn. The clearest separation was observed for Na, which presented systematically higher concentrations in rice samples originated from the east part of Rio Grande do Sul, i.e. close to the Atlantic Ocean. However other chemical elements also demonstrated to be useful for discriminating the geographic origin.

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Radiochemical separation of uranium and protactinium from neutron irradiated thorium.

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The aim of this work was elaboration of radiochemical scheme for separation micro-amounts of uranium and protactinium from macro-amounts of Th, what can be helpful in nuclear power engineering for the analysing thorium-uranium fuel cycle as well as IV generation nuclear reactors. Naturally abundant isotope of thorium, 232Th is the fertile material. In the nuclear reactor, 232Th is transmuted into the fissile artificial uranium isotope 233U.. Elaborated separation procedure is based on extraction chromatography, which combines the selectivity of liquid-liquid extraction with the rapidity and quantitativity of chromatographic methods. In first step, Pa is quantitatively isolated from micro-amount of U and macro-amount of 232Th on the column filled with TOPO supported on hydrophobic sorbent. Next, effluent containing thorium and U traces is loaded onto the column filled with quaternary aliphatic amine and an elution is conducted with a mixture of nitric and hydrofluoric acids with an addition of aluminium nitrate and H2O. Uranium is selectively eluted with first 5-10 mL, whereas Th is eluted in next 35-40 mL.

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Radiochemical separation of arsenic from selenium and its potential usage in generator isotope production.

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The potential usage of arsenic isotopes for nuclear medicine has been reported recently. One of the way for obtaining appropriate radioarsenic species is using radionuclide generator, where As is formed by the radioactive decay, eg. 72Se 72As. A new radiochemical separation scheme based on extraction chromatography for isolation As from Se is presented. The distribution coefficients of As and Se on prepared sorbents (selected aromatic o-diamines supported on polystyrene adsorbents) were determined in order to find the best condition for separation of both elements. Batch experiments were verified by column studies. Elaborated radiochemical separation scheme insures high selectivity and radionuclide purity of separated arsenic fraction, whereas examined sorbents have been found to have a very high selectivity with reference to selenium (IV). Arsenic is easily eluted by diluted HCl and NaCl solutions. The present separation scheme based on extraction chromatography, which could be used to designing a new 72Se/72As generator.

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Mineral Nutrients in Brazilian Commercial Dog Foods

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Mineral nutrients are essential chemical elements for a normal development of dogs. There are two groups of mineral nutrients required by dogs, macro minerals (Ca, P, K, Na, Cl and Mg) and trace minerals (Fe, Cu, Zn, Mn, Se, I). The body cannot synthesize any of them, thus dietary supplementation in proper quantity is crucial. Oversupplementation of one element may result in the deficiency of another, due to the interaction among minerals in the body. Either high or low level of a particular mineral in the diet can be harmful to dog's health, especially for growing puppies. Commercial dog foods must have balanced levels of mineral nutrients, which should follow regulations as that of the Association of American Feed Control Officials (AAFCO). In this context, a comprehensive sampling of dry dog food for puppies and adults of various brands was performed in the local market of Piracicaba city, Brazil, for evaluating the adequacy of mineral nutrients. Instrumental neutron activation analysis (INAA) was chosen to assess the chemical composition of the dog food, using two irradiations for determining both short and long-lived radionuclides. The moisture content of samples ranged from 5% to 12% in compliance with the values required by the Brazilian legislation for dry dog food. INAA was a suitable analytical tool for characterizing the profile of mineral nutrients in dog food, allowing the determination of Ca, P, K, Na, Cl, Mg, Fe, Cu, Mn, Zn, I and Se. In general, all concentrations of mineral nutrients were within the permissible limits established by AAFCO for growing puppies and adults, with some slight deviations. Therefore, one can consider the Brazilian commercial dog foods as having good quality in terms of mineral nutrients.

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A new PTS for short-time neutron activation analysis

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A fully-automatic pneumatic transfer system (PTS) for short-time neutron activation analysis (STNAA) is constructed to utilize two irradiation positions of a TRIGA Mark-II research reactor. The system consists of a vertical in-core irradiation terminal and a terminal for a radial radiation beam tube. Both terminals were constructed for a sample capsule of 3.5 ml. The transfer time of the irradiated capsule (4g) for a distance of 30 meters is found to be less than 3 seconds using pressurized air (3 bars). The Irradiation position of the sample capsule inside the beam tube is set at an angle of 45o and can be equipped with a moveable Cd filter for thermal/epithermal irradiations. The system is equipped with two sample changers for automatic analysis of un-irradiated and pre-irradiated samples for their short- or long-lived nuclides. A software package for the system is developed using two different codes. The first is based on PLC-code to control the irradiation procedures while the second is based on a Delphi to manage the measuring procedures with one of two digital spectrometers (Genie and Dspec). The software package manages the complete analysis procedure for reliable hardware/software control of the pneumatic- and the counting systems. The counting chamber is fabricated from Plexiglas and equipped with a linear motor to set the sample automatically at one of 4 geometry-positions, according to the activity of the analyzed sample.

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Natural radionuclides levels in spices and medicinal plants by gamma spectrometry

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This work is a contribution to the valorization of nuclear technics for environmental monitoring, mainly natural radioactivity in edible vegetation. The activities of natural radionuclides in major groups of widely used spices and medicinal plants in Tunisia have been investigated. In this aim, 18 selected samples imported from different countries were purchased in the dried form of roots, leaves, seeds, barks, flowers and fruits from local herbalists and farmers. Activity concentrations in all samples were measured by gamma spectrometry.
Specific activity concentrations measured in the considered samples show different levels in 40K and 238U. These radioanalytical results of 40K showed different levels activity concentrations ranging from 168.72 to 1154.1 Bq/kg. They are about 168.72 Bq/kg, 985.64 Bq/kg, 480.27 Bq/kg, 606.15 Bq/kg, 168.72 Bq/kg and 268.02 to 871.4 Bq/kg respectively for fruits, roots, flowers, leaves, barks and seeds. The activity of this nuclide is important in comparison to other radionuclides, which is due to the natural abundance of this essential nutrient in the soil and its high absorption by plants. Determined concentrations of 238U ranged from 11.02 Bq/kg and 123.18 Bq/kg. The highest concentrations were obtained for red pepper and fennel characterized with activity concentrations of 123.18 Bq/kg and 103.04 Bq/kg respectively.
According to this study, it was concluded that many factors such us plants composition, mobility of the radionuclides, climate and depth distribution of roots control the transfer and uptake of radionuclides from soil into plants.

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Detection of irradiated foods using TL, ESR and GC/MS

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Food irradiation technique is extremely effective at reducing food-borne illness as well as losses caused by pathogenic microorganisms. However, there is a need to detect the use of food irradiation to ensure that food is labeled correctly. In present, several methods are available to detect irradiated food. Among them, thermoluminescence(TL), electron spin resonance(ESR) and gas chromatography/mass spectrometry(GC/MS) detection are the leading techniques. The aim of this study is to set up applicability for foods which are not allowed to be irradiated in Korea. The groups of dried fruits and seeds were collected. The results showed that it is possible to apply TL method to all foods with mineral. In ESR method, food containing sugar showed radiation-induced crystalline sugar radicals, but the others did not exhibit radiation-induced radicals. In case of seeds, GC/MS method was applied and radiation-induced hydrocarbons were observed. According to the conclusion based on our experimental results, TL, ESR and GC/MS methods were successfully applied to examine irradiated and non-irradiated foods.

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The air-water partitioning of radon in groundwater contaminated by BTEX

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It is well known that organic solvent, such as benzene and toluene, has the highest level of solubility for radon, being approximately 50 times higher than the solubility of radon in water at the same environmental conditions such as temperature and pressure. The inhomogeneous distribution of radon between water and organic compounds allows for utilizing naturally occurring radon as aqueous tracer for the assessment of residual organic contamination of aquifers. In the present work, the air-water partitioning coefficient of radon in water contaminated by organic compounds were measured by a modified equilibrium partitioning in closed system (MEPICS) technique using a liquid scintillation counter (LSC). The organic compounds were benzene, toluene, ethyl benzene and xylene (BTEX). A radioactive standard solution (SRM4967, NIST) of radium (Ra-226) was used as a radon source of groundwater. The MEPICS method was applied to measure the partitioning coefficient of radon in pure water and an artificial water containing 30μg/L BTEX at 25 °C.

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Geochemically anomalous phonolites from Lusatian Mountains, Czech Republic: Possible source materials and processes of their origin

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The study has been aimed at elucidation of source materials and processes leading to formation of anomalous phonolites from the Lusatian Mountains (Bohemian Massif, Czech Republic). For a detailed geochemical study of the phonolites, various modes of instrumental neutron (short and long time irradiation, including the epithermal and fast neutron mode) and photon activation analyses were utilized. Combination of these modes allowed assaying up to 48 major and trace elements. Strontium and neodymium isotopic compositions were determined by TIMS.

Phonolites represent the subvolcanic (hypabyssal) rocks, which solidified from magma at relatively low temperature and consolidated in shallow depths in the upper bed. They are mostly associated with Tertiary interplate magmatism, but can originate also from differentiation of the upper mantle under the oceanic crust. Phonolites are strongly depleted by compatible elements as Mg, Cr, Fe, Co, Ni, Sr, whereas Na, K, Rb, and Al are enriched.

The origin of the studied phonolites of ca. 30 Ma age is associated with the Lusatian Fault. Compared to phonolites from the Eger Rift in the Bohemian Massif, contents of incompatible elements Zr, Hf, Nb, Ta, Th, U, light rare earth elements, F and Cl in the Lusatian Mountains phonolites reach extremely high values. Their 143Nd/144Nd isotope ratio is monotonous, corresponding to the upper mantle, whereas the 87Sr/86Sr ratios reach extreme values up to 0.8605.

The geochemical and isotopic data indicate that the anomalous phonolites may have originated mainly from partial melting of lower crust of tonalite-monzonite character or metamorphic rocks – shales with high contents of, e.g., micas. Participation of matter from the differentiated upper mantle is negligible. The high content of incompatible elements is probably associated, besides nepheline, sanidine and other foids, with accessory minerals such as zircon or titanite.

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Determination of Mineral Contents in Korean Domestic Unpolished Rice and Bean Samples by Neutron Activation Analysis

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As scientists have focused their researches on the health impacts caused by mineral nutrient deficiencies and hazardous elements, public concern regarding mineral intake from dietary food is rising. For this reason, the dietary habit of Koreans has shifted from white rice to more nutritious rice like unpolished rice and rice mixed with beans. The objectives of this study were to determine the mineral contents in unpolished rice and bean samples and to compare the level of mineral contents between the analyzed samples. Four kinds of unpolished rice and three kinds of beans were chosen as the target samples and seventeen mineral contents, Al, As, Br, Ca, Cl, Co, Cr, Cs, Cu, Fe, K, Mg, Mn, Na, Rb, Se, and Zn were determined by a neutron activation analysis. K shows the highest values among the analyzed elements from unpolished rice and bean samples. Only As in the unpolished rice samples shows higher contents than that in the bean samples. Ca, Cu, Fe and K in the unpolished rice are 8-times higher than those in the beans. Mg and Zn in the unpolished rice are slightly higher than those in the beans. Additionally, these results were compared with mineral contents in white rice

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Non-destructive and quantitative multi-elemental analysis by muonic X-ray spectroscopy for archeological bronze samples

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Quantitative elemental analysis is one of the most important themes in the field of natural science. For example, in the field of archeology, determination of elemental composition provides useful information to understanding the history and propagation of technology and culture. The archeological samples are highly valuable and the surface of these samples is usually oxidized or coated. Therefore, it is essential to get elemental composition deeply inside of the sample without destruction, however, such an analysis method is very limited.

In this paper, we report the development of a non-destructive, position-sensitive and quantitative multi-elemental analysis method for bulk samples by characteristic muonic X-ray measurements. When a negative muon that is one of the elementally particles is stopped in material, the muon is captured on a nucleus and muonic atom is formed. A muonic atom is an atomic system that has one negatively charged muon instead of an atomic electron. After formation of the muonic atom, the captured muon immediately de-excites to lower muon atomic levels through muon characteristic X-ray emissions. Because the mass of the muon is 207 times larger than that of the electron, characteristic muonic X-ray is much harder than electronic X-ray and muonic X-rays even from inside of a bulk sample are detectable.

In this study, we determined the elemental composition of old Japanese coins (tempo-tsuho) and old Chinese mirror (seiun-kyo). Muon irradiation experiments were performed in J-PARC (Japan) and Rutherford Appleton Laboratory (UK). Characteristic muonic X-rays were measured by high purity germanium detectors. We also performed muon irradiation for standard bronze samples to determine relation between characteristic muonic X-ray intensity and elemental composition. The details will be discussed in our presentation.

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Radiometric quantification of type 1 iodothyronine 5'-deiodinase activity in human white adipose tissue

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White adipose tissue (WAT) represents an important target for thyroid hormones (TH), which are known to modulate adipose tissue metabolism and differentiation of adipocytes. However, relatively little is known about TH metabolism in WAT. Recently, we determined higher activities of the key enzyme of TH metabolism, type 1 iodothyronine 5'-deiodinase (D1), in obese compared to lean control mice. Moreover, D1 activity in murine WAT could be stimulated by leptin, a major adipokine secreted in enhanced rate from hypertrophic adipose tissue. Based on these results, we aimed in the present study to measure activities of the three known iodothyronine deiodinases (IDs) in WAT of humans, and to characterize their possible association with obesity. Omental (OM) and subcutaneous (SC) adipose tissue samples were obtained during elective surgical procedures from a cohort of 70 human subjects with a body mass index (BMI) between 20 and 68 kg/m2. In a randomly selected subpopulation of 19 individuals, IDs activities of types 1 (D1), 2 (D2) and 3 (D3) were estimated in both OM and SC fat depots. IDs activities were measured in sub-mitochondrial supernatant fractions prepared from frozen WAT samples (10-40 ☒g protein in a final volume of 40 ☒l) using our recently elaborated radiometric enzyme assays. Specific D2 and D3 enzyme activities were close to the detection limits and there were no apparent differences in these activities between obese and non-obese subjects. On the contrary, activity of D1 could be quantified and it was significantly higher in both OM (ca. four-fold, P = 0.010) and SC (ca. eight-fold, P = 0.004) fat of obese when compared with non-obese individuals. Indeed, D1 activity correlated with BMI in both OM and SC fat depots. In conclusion, we have demonstrated for the first time in humans that (i) activities of enzymes involved in TH metabolism in human WAT differed in subjects according to the degree of adiposity; and that (ii) D1 was increased in hypertrophic adipose tissue (in obese subjects). Support from the Academy of Sciences of the Czech Republic (Project No. AV0Z50110509) and from the Czech Science Foundation (GACR Grant No. 304/08/0256) is acknowledged.

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Development of diet-induced obesity in the rat, followed by radioanalytical methods

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High-fat-feeding induction and development of obesity was followed in male C57BL/6J mice maintained on a special high-fat (HF) diet, in comparison with the same animals maintained on a standard low-fat (LF) diet. In parallel to several morphological parameters, changes in the thyroid status of the animals were followed. Serum total thyroxine (T4) and 3,5,3'-triiodothyronine (T3) concentrations were determined using radioimmunoanalytical kits. Moreover, with the aid of our newly developed radiometric enzyme assay, we measured changes in the enzyme activity of type 3 iodothyronine deiodinase (D3) in several depots of murine white adipose tissue (WAT). D3 is the key enzyme in the metabolism of thyroid hormones, converting biologically most active T3 and prohormone T4 into inactive metabolites (3,3'-diiodothyronine, T2 and 3,3',5'-triiodothyronine, reverse T3, respectively). HF-diet feeding resulted in an increased size of adipocytes and in a significantly higher weight of both epididymal-visceral and dorsolumbar-subcutaneous fat depots. Total T4 and total T3 plasma levels were significantly elevated in mice fed HF-diet, in comparison with mice maintained on LF-diet. Development of HF-diet-induced obesity in the mice was associated with an enhancement of D3 activity in WAT. After maintaining the mice on HF-diet for two weeks, the D3 activity was significantly increased in their subcutaneous fat (142 ± 20 vs. 85 ± 12 fmol T2/h/mg protein) but not so much in epididymal fat. In conclusion, HF-diet-induced obesity in mice was associated with proliferation and differentiation of WAT and stimulation of thyroid hormones metabolism in WAT. Support from the Academy of Sciences of the Czech Republic (Project No. AV0Z50110509) and from the Czech Science Foundation (GACR Grant No. 304/08/0256) is acknowledged.

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Mineral Elements Determination in Medicinal Plants

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The demand for herbal medicines is growing worldwide. According to data from the World Health Organization, approximately 80% of world population has resorted to the benefits of certain herbs with therapeutic action popularly recognized. The determination of major, minor and trace elements and the research of metabolic processes and their impacts on human health is of great importance due to the growth of environmental pollution that directly affects the plants and therefore the phytoterapics. Therefore, the objective of this paper was to determine the content of inorganic constituents in herbal medicine: moisture, inorganic ash, total ash, and the elements As, Ba, Br, Ca, Cs, Co, Cr, Fe, Hf, K, Na, Rb, Sb, Sc, Se, Ta, Th, U, Zn and Zr by neutron activation analysis in medicinal plants in order to verify the quality of products as well as their correlation with reference standards.
This study presents the results obtained for 59 medicinal plants commonly used in Brazil. Impurities were found in levels up 50% of the sample. The moisture levels varied from 0,1 to 12%. The concentrations obtained varied in a wide range for almost all the determined elements and the variation coefficient ranged from 50 to 245%. Good agreement was found between the results obtained in this work and the levels of trace elements reported in literature although high values for elements such as Ca, Ba, Cr, Fe and Zn were found in some samples.

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Validation of the method for Ni determination in NPP evaporator concentrates

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In this work a procedure developed for direct determination of radionickel in boric acid containg evaporator concentrate generated at VVER nuclear power plants has been validated for repeatability. For nickel separation, this method uses the composite material PAN-DMG (dimethylglyoxime incorporated in porous beads of polyacrylonitrile) to selectively bind 59+63Ni from the above mentioned concentrate. The PAN-DMG resin has been prepared by methods developed at our Department of nuclear chemistry at the Czech Technical University in Prague [1]. The method of 59,63Ni separation on column filled with PAN-DMG from real boric acid concentrates, that was validated in this work, has been developed in the thesis of Fišera [2]. In this work the influence of different uncertainty sources on the repeatability of this method has been studied. The repeatabilities of nickel separation, sample preparation, sample measurement, was investigated in this work. In addition, influence of the type of scintillation cocktails used on the measurement was determined. The results obtained showed that as well as whole method also the particular parts of the methods are repeatable.
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A preliminary study of prompt gamma-ray activation analysis using pulsed neutron at J-PARC / ANNRI

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The Accurate Neutron-Nucleus Reaction Measurement Instrument (ANNRI) has been constructed at the beamline No. 04 at the Material and Life Science Experimental Facility (MLF) of the Japan Proton Accelerator Research Complex (J-PARC). ANNRI has been used for the measurement of nuclear cross-section data, nuclear astrophysics and prompt gamma-ray analysis (PGA). MLF is a very intensive pulsed neutron facility, and expected to reach 1 MW proton beam power in the near future. The germanium detector-array, which consists of two cluster-Ge detectors, eight coaxial-Ge detectors and BGO Compton suppression detectors, was installed at the flight length of 21.5m in ANNRI. It is designed to provide high gamma ray energy resolution and high detection efficiency. The time resolution of Ge detector is poor but this is not a disadvantage because of the proton beam pulse width of 0.1 micro sec. We have developed a time-of-flight prompt gamma-ray analysis combined with gamma-ray coincidence technique. Preliminary experiments of BCR CRM 680 and 681 (polyethylene reference material) etc. were made in ANNRI. The power of the proton beam was approximately 200 kW and the repetition of the neutron beam was 25 Hz. In this study, the background spectra, the data acquisition dead time and the effect of the self-shield of neutron flux, which depend on the neutron energy, have been measured and evaluated. These are compared with the results of the experiments of multiple prompt gamma-ray analysis at Japan Research Reactor-3, and the similarity and difference are discussed.

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Biodistribution of Gadolinium-Based Contrast Agent, and Concentration of Trace Elements in Normal and Nephrectomized Mice

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INTRODUCTION: Gadolinium (Gd)-based contrast agents have been used in medical MR-imaging. However, human nephrogenic systemic fibrosis (NSF) is reported in Gd-based contrast agent received patients with severe renal insufficiency. A depletion of endogenous Zn ion may contribute in the development of NSF. Gadolinium has fairly high sensitivity for neutron activation analysis (NAA) and it is useful to know Gd behavior in biological samples as an activable tracer. In general, NAA technique also quantitates essential trace elements in vivo such as Zn, Mn. In this study, we measured distribution of Gd and concentration of some minerals in selected tissue of four experimental conditioned mice using of NAA.

EXPERIMENTS: Twenty male 8-week-old ICR strain mice were used in this study. Ten mice were housed as normal, and another ten mice were partially nephrectomized. Five mice in each group was administered 2.5 mmol Gd/kg body weight of Gd-based contrast agent intravenously. Two days after administration, mice were sacrificed and dissected. The blood, femur, kidney, and liver were excised and weighted. The samples were freeze-dried and sealed into polyethylene bags for NAA. The sealed samples, reference materials, and standard of Gd were irradiated by neutron at Pn-2 site of Kyoto University Research Reactor Institute. The distribution of Gd and concentration of minerals were determined by using gamma-ray spectrometry.

RESULTS: The nephrectomized mice showed high Gd retention in each tissue compared to normal mice. The kidney showed high Gd retention behaviors due to normal process of excretion of Gd. Skeletal Gd distribution of nephrectomized mice were significantly higher than that of normal mice. Manganese concentrations in kidney of model mice decreased relative to normal mice. On the other hand, Mn concentrations in liver were comparable during four groups. Zinc concentration in the selected tissue showed no difference during four groups of mice.

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Application of medium-energy proton beam from AIC-144 cyclotron in biological and environmental studies

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Activation analysis is a well known analytical technique used for performing both qualitative and quantitative analysis of major, minor and trace elements in a given sample, without or with chemical separation. Application of protons with the energy higher than 30 MeV in activation analysis considerably broaden the spectra of possible reaction channels (possibility of (p, xn) reactions, presence of the secondary neutrons which can also interact with a studied sample), which is always associated with some difficulties in the identification of activation products. This must be carefully examined in order to eliminate the existing interferences. AIC-144 cyclotron at the Institute of Nuclear Physics Polish Academy of Science can accelerate proton up to the energy of 60 MeV while the beam intensity does not exceed 50 nA (which is suitable for the radiotherapy of eye melanoma). Since 2010 we have studied several types of biological and environmental samples trying to check whether a proton beam with such intensity and energy can be used in activation analysis. As a result we have selected the elements which are possible to be determined under the above-mentioned experimental conditions. The rotating target holder enabled us to ensure the same irradiation conditions for samples and standard solutions.

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Sequential separation of ultra-trace U, Th, Pb, and lanthanides with a single anion-exchange column

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Abundance and isotopic ratios of trace uranium (U), thorium (Th), lead (Pb) and lanthanides in environmental samples play a key role to investigate features of the samples. In the environmental samples such as rocks, soils, and airborne dusts, trace U, Th, Pb, and lanthanides are contained with major elements such as Na, K and Fe. These major elements and the polyatomic ions which originated from the co-existing elements affect accuracy of analytical results measured by ICP-MS. The analytes, therefore, should be separated from the interfering elements. The authors developed the technique for sequential separation of U, Th, Pb, and the lanthanides using a single anion-exchange column and mixed media consisting of hydrochloric acid, nitric acid, acetic acid, and hydrofluoric acid. With a single anion-exchange column, sequential separation of 50 ng each of the elements of interest in an ICP-MS calibration solution was successfully carried out. The recovery yields of these elements were more than 95%. Alkali metals, alkali earth metals, and Fe were eliminated from these elements. This sequential separation technique was applied to the author's own simple automatic system. The system consists of a tiny anion-exchange column, switching valves controlled by a PC, and a gas bottle to pressurize the eluents. The separation of elements of interest was optimized for several parameters including particle size, column length, and flow rate of eluents. The elements of interest were completely separated within 6 hours by use of this system.

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Experimental Performance Evaluation of a Compton Suppression System for Neutron Activation Analysis by Using a Gamma-ray Source and Standard Reference Materials

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A Compton suppression system (CSS) was implemented at a neutron activation analysis laboratory of the Korea Atomic Energy Research Institute (KAERI) in 2009 and its performance was evaluated experimentally by the measurement of a gamma-ray emitting source and of detectable nuclides created by neutron activation with the NIST standard reference materials (SRMs). Four geological and five biological SRMs were chosen and irradiated by using an NAA#1 irradiation hole at the HANARO research reactor. A gamma-ray spectrum with normal mode and anti-coincidence mode were acquired at the same time, and advantage factors of CSS for each nuclide detected were calculated on the basis of signal-to-noise ratio. 531 keV of Nd-147 shows the highest advantage factor, 3.30, from geological samples, and 559 keV of As-76 shows the highest advantage factor, 2.36, from biological samples. The mean AF values for the nuclides detected from four geological and five biological SRMs were 1.99 and 1.63, respectively.

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NukWik – A Tool for Collaboration and Sharing Teaching Material in Radiochemistry

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Many institutions across Europe (and elsewhere) teach radiochemistry, more or less with the same content. At most institutions, the teaching material has been created and developed in-house through many years and frequently through many generations of teachers. In most cases, the material is not publicly available. However, from time to time institutions have to create new courses or at least update existing ones, which can be a large and time-consuming undertaking. For radiochemistry teaching, which in many places is only taught to small groups of students, justifying the personnel and other costs of developing teaching material and methods can be prohibitively difficult, and will frequently be done by idealistic individuals on their own time.

As part of the CINCH project [1] for developing teaching and training in Radiochemistry in Europe, we have therefore developed an open platform for sharing teaching material and even active collaboration across institute/university borders. The platform is run on a wiki engine and is called NukWik [2]. The best known example of a wiki is Wikipedia [3].

In brief, a wiki can be described as an editable web site. To edit a wiki there is no need to know complicated computer code or similar (e.g. HTML), in most cases it is done in a simple and easy to understand web-page interface. Content can be added or edited by any member of the wiki user-group provided an internet connection is available. Content is coordinated by links between the pages in addition to different categories. Categorising the wiki-pages is very important, as this is one of the most common ways to seek out relevant information. Furthermore, each wiki-page has an associated discussions page. This can be used by teachers to discuss how to improve and use the material, e.g. a specific laboratory exercise.

With NukWik, teachers can share and collaborate on anything from a given calculation exercise to complete courses, including extensive laboratory exercises. Institutions will of course still "own" their own courses and students, but teaching material and experience can be shared. In this way one can make better courses with updated and relevant content without single-handedly taking the full burden of time and resource costs.

Of course, NukWik only works if those using it are willing to freely share their material and experience. If you are such a person, please do not hesitate to log on and/or contact us!

References:
[1] CINCH is a 7th Framework Program of the European Commission (Euratom), project web-page is located at http://cinch-project.eu/index.php
[2] https://wiki.uio.no/mn/safe/nukwik/index.php/NukWik
[3] http://en.wikipedia.org/wiki/Main_Page

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Fluorescent Imaging of the Radiation Dose Surrounding an Iridium-192 Seed Used in Brachytherapy

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The energy transferred to the medium surrounding a small seed of a radioactive isotope decreases steeply with distance from the seed and depends on the nature of the particulate and/or electromagnetic radiation emanating from it. This highly-localized dose distribution is used to effect in brachytherapy for the radiotherapeutic treatment of malignant tumours. Accurate monitoring of such a steeply-varying dose distribution requires ideally a dosimetric technique capable of three-dimensional, sub-millimetre spatial resolution. We have developed such a technique that is based on a gel medium that becomes fluorescent on exposure to ionizing radiation with the intensity of the fluorescence proportional to the local radiation dose. We illustrate the method with photographic images of the fluorescence surrounding a high dose rate (HDR) seed of iridium-192. The images, with a spatial resolution of better than 0.1 mm, were made in situ and could be recorded in real time during the course of irradiation using time-lapse or video recording modes. The gel is close to tissue-equivalent and its radio-fluorogenic property is based on the radiolytic formation of reactive free-radicals. The measurements are therefore directly relevant to the radio-biological processes responsible for the destruction of malignant tissue. This method of real-time, 3D dosimetry should expedite the research, development and application of radio-nuclides for radiotherapy and provide a visual image that could be of use in the education and training of clinical personnel.

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66Ga-labeling of DOTA-conjugated cyclic RGDfK dimer for αvβ3 integrin overexpression tumors

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Angiogenesis is an essential process in solid tumours growing beyond 2 to 3 mm3, since diffusion is no longer sufficient to supply the tissue with oxygen and nutrients. Integrins αvβ3 have been shown to play an important role in a series of pathological processes including angiogenesis and tumour cell metastasis. It has also been shown that peptides based on the Arg-Gly-Asp (RGD) sequence have a high affinity and selectivity for αvβ3 integrins. The aim of this research is to prepare and evaluate 66Ga-DOTA-Glu-[cyclo(Arg-Gly-Asp-D-Phe-Lys)]2 (66Ga-DOTA-E-[c(RGDfK)]2), as potential diagnosis agent for the early and specific detection of cancers overexpressing αvβ3, and evaluate its potential as possible agent for therapy. Gallium-66 was produced on a 11 MeV cyclotron via the 66Zn(p,n)66Ga reaction and radiochemical separation was performed by ion exchange chromatography using a AG 50W X-4 cation exchange column. For radiolabelling 30 µl of the peptide conjugated solution (400 ug/ml, H2O with 1% EtOH), 25 µl 1 M HEPES, pH 7.0 and 25 µl 0.25 M NH4OAc, an pH 5.5, were mixed with 66Ga stock solution (50 µl 0.1M HCl) and incubated for 20 min at 95°C. When needed, labelled peptides were purified by SPE on an Oasis HLB cartridge, product eluted with a small volume of 25% EtOH and diluted to <5% EtOH in PBS. In vivo studies were performed on a Focus 120 microPET System using C6 and U87MG xenografts in nude mice.

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NORMA: A new PGAI-NT setup at the Budapest Research Reactor

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Efforts are being made at laboratories worldwide to develop Prompt Gamma Activation Analysis (PGAA) towards a position-sensitive technique. It was proven earlier that the complete scanning with a few-mm-resolution is only practical on small objects due to constrains of experiment time and neutron flux. A feasible alternative is the combination of neutron radiography with prompt gamma activation analysis. Radiography, or even a full tomography of the complex sample, as a first step, can be completed in minutes, but often provides enough information to set up regions of interests inside the sample. The detailed element analysis by PGAA is carried out then only at these spots, saving substantial beam time. This novel combination of methods is best used for real samples which consist of a few homogeneous parts.

The first ever pilot instrument for this purpose was installed at the Budapest Research Reactor in 2007 and has been presented at the NRC7 conference. The technique proved its usefulness and raised enough interest in the user community to continue the efforts. Based on these experiences the first permanent radiography-driven PGAI facility, NORMA (Neutron Optics and Radiation Measurement for element Analysis) was constructed and put into operation in the first months of 2012.

Its specifications supersede the pilot setup in every aspect. The sample chamber has now dimensions of 20×20×20 cm^3. By removing one or more side panels, larger objects up to 5 kg weight could be analyzed (such as a sword, vase, stones, etc.). Samples can also be loaded to the chamber manually from the top side. The positioning table has a nominal travel distance of 200 mm. The gamma radiation is detected with a Compton suppressed system, that consists of a central Canberra GR2318/S HPGe detector surrounded by a Bismuth Germanate (BGO) scintillator made by Scionix. The cylindrical and exchangeable lead collimators can be mounted into a socket of the 10-cm thick lead shielding. The gamma events are collected with a Canberra DSP-2060 digital signal processor, in anti-Compton mode. The imaging system comprises a 100 um thick Li-6/ZnS scintillator, a silver-free quartz mirror set in 45 degree to the neutron beam and a cooled ANDOR iKon-M CCD camera (16-bit ADC and 1024×1024 pixel resolution), mounted to a light tight aluminum housing. Integrated data acquisition software operates the moving table, the gamma-ray spectrometer and the camera.

The commissioning has just recently been completed, and the facility became open to the international user community through the EU FP7 NMI3, CHARISMA and EFNUDAT access programs. The first results of this non-destructive technique will be presented here from various fields of applications, such as archaeometry, safeguards and material science.

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SYNTHESIS AND X-RAY STUDY OF RADIUM METAPLUMBATE

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The report presents experimental results on the synthesis and properties study of radium- and lead-based mixed oxide. Samples for examination were produced by high-temperature treatment of mixed $RaCO_3/PbCO_3$. A mixture of 2.0 mg of $RaCO_3$ and 5.0 mg of $PbCO_3$ was calcinated at 800°C for 8 hours in a stainless steel crucible. Resulted sample was studied by X-ray difractometry. In the X-ray pattern 15 reflexes of 43 registered ones were the strongest and referred to the phase being an analog of $BaPbO_3$ with a cubic perovskite structure.

Taking into account potential contamination of radium plumbate with corrosion product coming from steel crucible, the experiment was repeated with the use of a platinum crucible. Sample of radium nitrate used in this run was preliminary purified by cation-exchange chromatography: the barium content being decreased to 100 □g and the total amount of impurities Ca, Fe, Ba, Al, B, Si were below 76 □g.

During the second experiment, 2.9 mg of $RaCO_3$ and 4.6 mg of $PbCO_3$ were calcinated at 800 °C for 20 hours in the platinum crucible. The interpreted X-ray pattern showed the strongest reflexes to belong to metal platinum that came to the sample as an impurity. The other group of intensive reflexes corresponds to the phase of cubic $RaPbO_3$. The X-ray pattern also demonstrated a group of 5 reflexes close in the location and relation of their intensity to $BaPtO_{2.38}$ phase reflexes. More probably, this group of reflexes corresponds to radium platinate $RaPtO_{3-x}$ produced while radium reacts with the crucible material.

Based on supposition of $RaPbO_3$ formation, the interplanar space values were calculated as well as crystalline lattice parameter (a=4.303 Å) and crystallographic density (ρ=10.0 g/cm3).

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The influence of iron on the efficiency of the 68Ga labeling of DOTATOC and simple colorimetric determination of iron

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Introduction

It is well known that iron ions have a strong impact on the labeling efficiency of DOTA-peptides with 68Ga. In a draft for a monograph of 68Ga-chloride for the European Pharmacopoeia (PHARMEUROPA, Vol. 23, No. 3, July 2011) a maximum concentration for iron is given with 10 µg/GBq. The concentration should be determined by atomic absorption spectrometry.
Easier to handle but high sensitive and specific colorimetric test systems would have many advantages for clinical production of radiopharmaceuticals. Moreover, the European Pharmacopoeia describes well known methods for colorimetric determination of the total heavy metal concentration in pharmaceuticals and starting materials.
In this work we quantify the influence of iron and investigate the practical application of a colorimetric test, which is able to determine lower iron concentrations than required.

Methods

For the labeling of DOTATOC the NaCl based method (Mueller et al. , Bioconjugate Chem., 2012, 23 (8), pp 1712−1717) was used. A stock solution of iron(III) was prepared by dissolving of iron(III)-chloride in 0.1 M HCl. Different amounts of iron were added into the reaction vessels prior to the addition of 68Ga and the pH of the reaction mixtures were adjusted with sodium acetate buffer.
After heating the ratio between peptide-bound and free 68Ga was determined by radio HPLC (RP-18 column; solvent A water, solvent B acetonitrile, gradient 5-95%, 0-15 min).
The concentration of iron was cross-checked by a colorimetric test system (VWR, Microquant Iron Test, method colorimetric, Ferrospectral, 0.1- 5 mg/ml Fe).

Results

We could impressively show that the colorimetric iron test, cross-checked by a self made iron standard, safely determines the iron concentration in the reaction mixture or the eluate. The sensitivity of this test is hundred times higher than required. If 50 µg DOTATOC were used, a content of iron up to 0.6 µg shows a successful labeling but already 2 µg iron leads to an incomplete labeling. In this case the concentration of free 68Ga in the final reaction mixture is higher than 10 %. The use of 40 µg DOTATOC and 2 µg iron in the reaction mixture makes a successful labeling impossible. The concentration of 68Ga in the final reaction mixture is then higher than 90 %. After addition of 6 µg iron and the use of 50 µg DOTATOC the final concentration of free 68Ga amounts 48%. A prior concentration step with the help of a cation exchanger cartridge collects also iron(III) and it can be also eluted completely with an eluent for 68Ga-chloride. The total amount of iron in the 68Ge/68Ga generator eluate reaches therefore into the reaction mixture and would influence the labeling.

Conclusion

A content of 10 µg iron in the 68Ge/68Ga generator eluate would prevent a successful labeling of DOTA conjugated peptides. The colorimetric determination of iron is particular well suited to detect iron incorporation in the eluate or in the reaction mixture. This test system is much easier to handle than an atom absorption spectrometer in the clinical practice. Furthermore we assume that the established colorimetric test on heavy metals for pharmaceuticals and starting materials as describe in the European Pharmacopoeia would

safely show toxicological concentrations on these cations.

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Separation of actinium-225 for nuclear medicine purposes from thorium targets irradiated by high energy protons

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Due to high linear energy transfer (LET) of about 100 keV/μm and low path of about 50-100 μm in biological tissue of alpha particles, alpha-emitters are promising for targeted radiotherapy of cancer. The number of potential alpha-emitters that could be used in medical applications is limited by the requirements of half-life, availability of these alpha-emitters and experimental difficulties in their separation. The most promising candidates for clinical applications are 211At, 212Bi, 213Bi, 225Ac. Among these alpha-emitters, 211At could be easily obtained by irradiation of bismuth by 28-29 MeV alpha-particles on a cyclotron. However its application is limited by the synthetic preparation of astatineorganic compounds and weakness of At-C chemical bond. Actinium-225 could be used either directly for preparation of radioimmunoconjugates or could be used as a parent radionuclide in 213Bi isotope generator. However wide application of 213Bi and 225Ac is limited by the low availability of these radionuclides. Several methods are proposed for 225Ac production while most frequently it is separated from mother 229Th or by irradiation of 226Ra with protons in a cyclotron. Both methods have some practical limitations to be applied routinely.
The aim of this investigation was to develop the separation method of 225Ac from natural thorium irradiated with protons with the energy of around 100 MeV at linear accelerator of Institute of Nuclear Research RAS.
Thorium foil was irradiated by protons with the energy of 104 MeV. The radionuclide composition was determined by gamma-spectrometry. Irradiated thorium target was dissolved in minimal volume of 6 M HCl upon heat. Thorium and some fission products were separated by solvent extraction by TPB in toluene (1:1) (twice) and by 0.15 M TOPO in toluene. In some experiments dissolution was done by 7 M HNO3 and extraction was also done from this solution. Aqueous phase containing actinium was evaporated near dryness and dissolved in 0.05 M HNO3. These solutions was transferred to the extraction chromatographic column filled with Ln Resin (Eichrom) After adding the sample to the column, it was sequentially washed by 0.05 M HNO3 and 3 M HNO3 solutions. Actinium and REE containing fraction were loaded to the extraction chromatographic column filled with TRU Resin (Eichrom). To determine the impurities of 227Ac in actinium fraction the alpha-spectrometry was employed. Other radionuclidic impurities were assessed by gamma ray spectrometry. It was shown that most significant impurity is 227Ac (less than 0.1% on EOB) but it could not interfere in application of the product in Ac/Bi generators.

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Fast beta-alpha-pile-up suppression electronics for super heavy element identification

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The synthesis [1] and chemical [2] investigation of new elements are one of the most fascinating tasks in nuclear physics and radiochemistry. Especially for the investigation of their chemical properties the unam¬biguous identification of the decay of their isotopes is a very important prerequisite. The time-re¬solved spectroscopic recording of alpha particles emitted during the decay of an isotope as well as its con¬secutive decaying descendents gives the highest possible confidence to assign the observed event chain to the decay of the isotope of interest. However, the formation of unwanted by-products in the used nuclear reactions leads to large amounts of unwanted events in energy regions, where the decay of the inves¬tigated isotopes or their daughter nuclei is expected. Especially the beta-decay of Bi-212 and the following alpha-decay of Po-212 (half-life 0.3 us) might disturb the measurement due to the consecutive emission of a high energetic beta- and alpha-particle. The sum of the kinetic energies of both particles may exceeds 10.5 MeV if both hit the same detector within a short time period of typically less then 1 us. Conventional spectroscopy electronics can not resolve the consecutive hits of the beta- and alpha-particle in time. This leads to so called pile-up pulse height amplification. Also the separation power of high acceptance physical mass sepa¬rators like the Berkeley Gas-filled Separator (BGS at LBNL, USA) or the Trans Actinide Separator and Chemistry Apparatus (TASCA at GSI, Germany) are not able to suppress all of these beam-like by-products. It was shown in [3] that such pile-up events contribute two orders of magnitude more to the event budget in the region between 8.9 MeV and 9.5 MeV than all other possible events in the course of experiments synthesized 114 at the TASCA separator.
A new and fast spectroscopic detection system PURECOLD was designed and build-up to distinguish pile-up signals from real pure alpha-events at the same energy. This setup is able to record each 2 us a single event with overall event rates up to 12000 per sec and per spectroscopic channel. The discrimination of pile-up signals is based on the sampling of the leading edge of the independently amplified fast timing signal using sixteen consecutive fast voltage comparators (switching time 150 ps) with a time resolution of 0.5 ns. The spectroscopic part of PURECOLD consists of two independently acting amplifier chains each connected to a 500000 sample per second 16-bit analog digital converter to process events from 0 MeV to 30 MeV and from 0 MeV to 300 MeV, respectively. Furthermore, this system has the capability to measure time-wise very short consecutive alpha-decays, which appears to be increasingly important for the dis¬covery of new elements and new isotopes due to the possibility to convert a second signal arriving within the conversion time of the first one using the second amplifier chain and analog digital converter.

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Advanced Fuels for Generation IV reactors: Reprocessing and Dissolution – ASGARD

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ASGARD is a Large Scale Integrated Project having as main objective the research on advanced/novel nuclear fuel fabrication and their respective reprocessing issues for Generation IV reactors.
The Strategic Energy Technology plan (SET-plan) of the European Commission identifies fission energy as an important contributor to meet long-term objectives for reduction of greenhouse gas emissions. Sustainability of nuclear power may be achieved by the introduction of fast neutron Generation IV reactors and their associated fuel cycle facilities, as described in the Strategic Research Agenda (SRA) of the Sustainable Nuclear Energy Technology Platform (SNE-TP).
The project seeks integration between reactor, fuel and recycling communities, which today is lacking. In some cases it results in discrepancies between the reactor design on one hand, and the technological feasibility of fabricating, dissolving and reprocessing the selected fuel on the other hand.
Oxide, Nitride and Carbide type of nuclear fuels are addressed with focus on dissolution, reprocessing and fabrication behavior.
It is an integrated effort of 16 institutions from 9 countries, and additional: Scientific Advisory Commission (4 representatives) and an Industrial Users Group (5 representatives).
The consortium consists on partners who have an extensive experience in research on fabrication of various kinds of fuels and in the recycling process. In addition, basic science laboratories participate with large capabilities in handling of highly active radioactive materials such as Chalmers University of Technology, Sweden as well as national research laboratories and also university laboratories with more limited radiochemical possibilities.
In order to ensure the know-how and appropriate training of the future generation, the project will allocate a large space to training and dissemination. Training sessions will consist on well selected lectures and hands-on training as well as summer schools. A travel fund is allocated for mobility of students and teachers and will be based on travel grant system with defined rules.
The main goal is to ensure the sustainability and safety of future nuclear power fleet.
It is funded by the 7th Framework Program (FP7) of the European Commission (Euratom).
It was officially launched on the 11th of January, 2012 and will continue for four years.

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Problems of determination of Tc-99 in soil and sediments

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The 99Tc is a long lived (T1/2=2.11×105 years) fission product. It is worldwide spread as remains of global fallout. It is also released from nuclear reprocessing factories and was present in nuclear accident fallout, like Chernobyl for instance. This is also a decay product decay of short lived (T1/2=6 h) 99mTc, the most popular nuclear medicine isotope. The large difference in half life time reduces the activity in huge scale: 1 GBq of parent isomer state produces 3 Bq of the daughter. fallout. Since the global fallout was formed as a sum of deposition from hundreds individual events in a course of years the resulting deposition pattern is supposed to be rather uniform. Chernobyl fallout is much more patchy.
Our project was devoted to the recognition of the presence of 99Tc in environment of Poland. Samples of peat, forest soil and litter, sewage and river sediments were subject of our analyses. The 99Tc is a pure beta emitter (Emax 300 keV) and thus it can be measured by LSC. Due to long half life time it is also possible to measure it by means of ICP MS. In course of our analyses both techniques were applied. After mineralization and Tc separation on TEVA column each sample was divided into two parts: one was counted using LSC, second was examined by means of ICP MS. The chemical recovery was monitored using 95mTc and gamma spectrometry but an attempt was done to replace it by stable renium.
The presentation will describe the difficulties which we faced in course of the project and the ways how we solved the problems. The results of analyses will be presented as well.

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OPENING LECTURE - Recent Advances in Superheavy Element Research

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With the recent synthesis of element 117, the 8th row of the Periodic Table of the Elements is complete. Also, the elements with atomic numbers 114 and 116 have now officially been named Flerovium and Livermorium by IUPAC. Significant progress has not only been achieved in synthesizing new elements, but also in their chemical characterization. In my contribution I will give an overview of the status of the experimental work on the attempted synthesis of elements beyond Z=118 and then review the gas-phase chemistry of Hassium, Copernicium, and Flerovium and compare the results with theoretical predictions. Then I will discuss latest developments concerning chemistry experiments with pre-separated heavy element beams, that hold promise to chemically investigate so far not studied elements in the Periodic Table, which leads to an outlook about the near and far future of superheavy element research.

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INVITED LECTURE - Spectroscopic methods for the heaviest nuclei

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A growing number of experiments is currently opening up the transfermium region of nuclei for detailed spectroscopic investigations [1,2]. In the deformed nuclei in the nobelium region this allows an identification and mapping of single particle orbitals closest to the top end of the nuclear chart.

Initial in-beam measurements in the region focussed on γ-ray spectroscopy of even-even nuclei (e.g. 252,254-No, 250-Fm), studying the ground-state yrast bands and allowing extraction of parameters such as the moments of inertia, and proving the deformed nature of these nuclei. More recently, attention has switched to odd-mass nuclei such as 253-No, 251-Md and 255-Lr, the latter being the heaviest nucleus so far studied in-beam. Rotational bands have been observed in all these nuclei. Non-yrast and K-isomeric states have recently been observed in 252,254-No and 250-Fm through the use of both in-beam and focal plane decay spectroscopy. The studies employed a calorimetric technique, whereby the summed energy from a cascade of conversion electrons is detected in a DSSSD detector and used as a "tag" for γ-rays detected in the various germanium detectors. These experiments have yielded data which can be used to determine the excitation energies and configurations of two-quasiparticle states in the region, and compared to the predictions of various theories.

An overview of the most recent results and the experimental techniques used will be presented and new experimental developments such as the SAGE spectrometer in Jyväskylä and the new TASISpec setup at will be discussed.

This work is supported by the UK STFC.

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INVITED LECTURE - The unique chemical and physical properties of the heaviest elements in the Periodic Table

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The unique role of the heaviest elements in chemical and physical sciences is discussed. With the actinide series (Z = 90 – 103) and the superactinide series (Z = 122 – 155), the heaviest elements have significantly shaped the architecture of the Periodic Table of the elements. Relativistic effects in the electron shells of the heaviest elements change the chemical properties in a given group in a non-linear fashion. Relativistically stabilized sub-shell closures give rise to a new category of elements in the Periodic Table: volatile metals. The prototype for this property is element 114 which, due to the relativistic stabilization of its 7s27p1/22 electron configuration, is volatile in its elementary state, but, in contrast to a noble gas, exhibits a marked metal-metal interaction with a gold surface at room temperature. Nuclear shell effects dominate the physical properties of the transuranium elements. These give rise to superdeformed shape isomers (fission isomers) in the actinides (U – Bk). Superheavy elements (Z ≥ 104) owe their existence solely to nuclear shell effects at N = 152, 162, and 184. At this time, a building lot is the location of the next spherical proton shell closure as there is evidence that the center of the “island of stability” is not at Z = 114. This needs urgently further theoretical and experimental efforts. The cross sections for the syntheses of elements 119 and 120 will give us important information on the “upper end of the Periodic Table of the elements”.

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INVITED LECTURE - Vacuum thermochromatography - prospective method for heaviest element studies

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The title technique for separating species with dissimilar adsorbability uses deeply evacuated columns with imposed negative longitudinal temperature gradient. Then the molecular flow, though convectionless, produces deposits peaking in individual temperature ranges. This fundamental mechanism - random walks from wall to wall - calls for rigorous Monte Carlo (MC) simulations, which require a minimum of assumptions, are very versatile in accounting for numerous parameters and conditions of real experiments, but are slow. Diffusional approximation of random walks with solution to the appropriate differential equation gives excellent agreement with MC. The calculations are faster for some basic conditions, but the versatility is much poorer. Meanwhile, the structure of the solution suggests a way to a semi theoretical explicit formula for the peak. The one presented here is negligibly inaccurate, reasonably versatile, and unmatched in the speed of the necessary calculations. It greatly enables evaluation of the experiments with a few radioactive atoms or labeled molecules, which is the case in today chemical studies of the superheavy elements. This is demonstrated by simulating experimental data of this kind for certain desorption energy and then, backward, evaluating the confidence intervals for the "experimental" value of the energy. The Bayesian approach is used; several caveats on hasty conclusions at poor statistics are emphasized. Some considerations about the real experiments are presented.

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INVITED LECTURE - Aqueous-phase chemistry of the heaviest elements

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Chemical characterization of the heaviest elements in aqueous-phase experiments is performed by a partition method with single atoms, e.g., liquid-liquid extraction, ion-exchange chromatography, and reversed-phase extraction chromatography. In these processes, the behavior of the heaviest elements is compared with that of its lighter homologues under strictly identical conditions. Several investigations have been carried out with automated rapid chemical separation apparatuses to characterize chemical properties of the heaviest elements. Typically interesting results recently obtained are that fluoride complex formation of element 104, Rf, is significantly different from that of the homologues, and that anionic fluoride complexation of element 105, Db, is considerably different from that of the group-5 homologue Ta while the behavior of Db is similar to that of the lighter homologue Nb.
Development of a new apparatus based on flow electrolytic column chromatography combined with ion-exchange separation has been recently carried out. Oxidation of element 102, No, has been successfully conducted based on an atom-at-a-time scale. This approach will lead to new frontiers of aqueous-phase chemistry of the heaviest elements; information on valence electronic structure of the heaviest elements through redox potentials will be obtained. Prospects for the future studies on redox properties of the heaviest elements will be briefly considered.

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ORAL PRESENTATION - The observation of a volatile compound formation with Po and Bi during experiments with superheavy elements

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Sophisticated gas-phase chromatography experiments with superheavy elements Cn, E113, and Fl are performed using noble gas atmospheres as carrier gases with only trace amounts of oxygen and water mainly to ensure the elemental state. Surprisingly, during these experiments the gas phase transport of volatile species of Po and Bi have been observed. Their deposition behaviour on gold surfaces have been measured. The decay characteristics of the observed isotopes showed that the observed transport is not related to any precursors such as Pb or Rn. The dew-point monitoring of the gas and the pretreatment of the carrier gas with hot Ta-getters changed the trace gas composition and was shown to influence the formation of these compounds dramatically. From these observations the formation of volatile hydride compounds was concluded, opening up an interesting chemical system for the investigation of the superheavy elements E115 and Lv.

The work is presented on behalf of the PSI-University of Bern-FLNR-ITE collaboration.

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ORAL PRESENTATION - New experiments to study properties of ^{268}Db produced in the ^{48}Ca + ^{243}Am reaction

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We conducted a new round of experiments to isolate and study chemical properties of 268Db, the end product of 288115 synthesized in a 48Ca + 243Am reaction. In contrast to previous experiments [1, 2] the reaction products passed through a gas filled recoil separator (DGFRS) and were stopped in a catcher (copper foil) which provided an additional separation from the actinides with a decontamination factor of 4 – 5 orders of magnitude. The chemical separation procedures have been developed for selective isolation of Db from the catcher using coprecipitation with LaF3 [3], ion-exchange and extraction chromatography from HF and HCl solutions. Altogether 8 spontaneous fission events were detected with a half-life of 23+13-6 hour. The cross-section for the production of 288115 was found to be 6+3.6-2.4 pb. This is in agreement with the previously reported data [1, 2, 4]. According to all previous experiments the half-life of spontaneously decaying nuclide 268Db was estimated to be 27.4+4.6-3.4 hour. Also the extraction behavior of Db together with those of the lighter group-5 elements Nb and Ta into UTEVA resin was investigated in 2 – 5.5 M HCl solutions by reversed-phase extraction chromatography. The obtained order of extraction Nb > Ta > Db suggests that stability of the chloride complex of Db is lower than those of Nb and Ta.

The work was supported by Russian Foundation for Fundamental Research (project code 11-03-12058-ofi-m).

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ORAL PRESENTATION - Superheavy element 114 is a volatile metal

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The experimental determination of chemical properties of element 114 (E114) is among the hottest topics in superheavy element research. A first experiment reported E114 to be highly volatile, and to form weak physisorption bonds with Au surfaces [1]. However, the large uncertainties of the measured adsorption enthalpy covered a wide range in volatility, which prevented the experiment from yielding an unambiguous answer concerning the chemical properties of E114. A noble gas-like behavior, representing a break in the trend in group 14 in the periodic table, would be in contradiction to many recent theoretical calculations, which predict a higher volatility and inertness compared to the lighter homolog Pb, but a stronger metallic behaviour compared to Cn [2-4].

We have performed a gas phase chemical study of E114 using a combination of the TransActinide Separator and Chemistry Apparatus (TASCA) to isolate single atoms of E114 [5], and the Cryo-Online Multidetector for Physics and Chemistry of Transactinides (COMPACT) [6], a gas chromoatography detector suitable for studying the interaction of single atoms with metallic Au surfaces. The setup allowed studying elements covering a broad range in volatility, from the non-volatile heavy metal Pb to the noble gas Rn, at a very low background level.

In our experiment, the volatility of five elements was studied: the two superheavy elements E114 and Cn (Z=112), their lighter homologs Pb and Hg, and the noble gas Rn. Two element 114 decay chains, one

from288114 and one from 289114, have been detected and indicate E114 to adsorb on Au surfaces at room temperature [7]. The interaction of element 114 with Au is at least as strong as that of Cn, in contradiction with a previous experiment [1]. Our results show element 114 to be the least reactive member of group 14, but still a metal.

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ORAL PRESENTATION - Exploration of the metallic character of astatine

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Astatine 211 is considered to be one of the most promising candidates for targeted alpha therapy (TAT)1, 2 and it is the subject of a wide research program in Nantes (France).
Very few data on the chemistry of astatine (At) are available. On the one hand, At is a rare element and it has only short half-life radioactive isotopes. On the other hand, it is an "invisible" element: the amount of At-211 produced allows working at ultra trace concentrations (typically 10-11 to 10-15 M) and no spectroscopic techniques can be used to estimate At characteristics at the molecular level. As a result, At chemistry is not well understood.
Based on these considerations, a research program has started in Nantes to explore the fundamental properties of At using a multi-disciplinary approach combining radiochemistry, analytical chemistry and molecular modelling competences. The object of this contribution is to present the main advances obtained during the last 8 years as regards especially to the particular metallic character of astatine. The methodology enabled us to define a Pourbaix diagram (Eh/pH diagram) for At in non-complexing acidic aqueous medium. We showed the existence of At-, and two stable At+ and AtO+ metallic forms of astatine.3, 4 This highlighted the metallic character of At by comparison with others halogens, as it was already proposed in the 60's.5 Our recent results on the chemical reactivity of AtO+ demonstrate the potentiality to form both coordination and covalent bondswith organic and inorganic ligands.6, 7

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ORAL PRESENTATION - RIKEN GARIS as a promising interface for superheavy element chemistry –Production of 261Rf, 262Db, and 265Sg for chemical studies using the GARIS gas-jet system–

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Chemical characterization of superheavy elements (SHEs, atomic numbers Z ≥ 104) is an extremely interesting and challenging subject in modern nuclear and radiochemistry. We have been developing a gas-jet transport system coupled to the RIKEN gas-filled recoil ion separator GARIS as a novel technique for SHE chemistry. This system is a promising approach for exploring new frontiers in SHE chemistry: (i) the background radioactivities of unwanted reaction products are strongly suppressed, (ii) the intense beam is absent in the gas-jet chamber and hence high gas-jet transport efficiency is achieved, and (iii) the beam-free condition also allows for investigations of new chemical systems. In this work, we investigated the performance of the system using 261Rf (Z = 104), 262Db (Z = 105), and 265Sg (Z = 106) produced in the 248Cm(18O,5n)261Rf, 248Cm(19F,5n)262Db, and 248Cm(22Ne,5n)265Sg reactions, respectively. The evaporation residues of interest were first separated in flight from the beam and the majority of the nuclear transfer products by GARIS and were guided to the gas-jet chamber at the focal plane of GARIS. The evaporation residues were thermalized in helium gas in the gas-jet chamber, were attached to KCl aerosol particles, and were transported through a Teflon capillary to a chemistry laboratory. Alpha and spontaneous fission decays of 261Rfa,b, 262Db, and 265Sga,b were then investigated with the rotating wheel apparatus MANON under extremely low background conditions. In the conference, productions and decay properties of those nuclides will be discussed in detail. A chemistry program using the GARIS gas-jet system will be also presented.

Session 4 - Reaction mechanisms and nuclear recoils, nuclear base spectroscopies, radiation geochronology, isotope effects / 196

OPENING LECTURE - Iron speciation in aqueous systems: the power of Mössbauer spectroscopy applied in frozen solutions

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Mössbauer Spectroscopy is technique basically used for the analysis of solid systems due to the neccessity of recoilless gamma emission and absorption. 57Fe offers very convenient conditions for Mössbauer measurements, and this is why iron containing systems ranging from alloys to minerals have been studied and reported in thousands of scientific papers, and Mössbauer Spectroscopy became a popular and powerful method.
Aqueous solutions of iron salts can only be studied in frozen state, which poses some experimental challenge, but this opens a new window to systems otherwise difficult to analyse.
Structure of solvated ions, species participating in complex equilibra, and numerous other problems can be solved by quick freezing of the solution studied.
The technique has been applied first by Attila Vertes in the late 1960s, and most common iron salts (nitrate, perchlorate, sulfate, chloride, etc.) have been thoroughly investigated.
Beyond the regular Mossbauer parameters, in solutions, paramagnetic spin relaxation can help distinguish between various species especially between monomeric species and possible bridged dimers or oligomers. Evaluation of Mössbauer parameters can reveal oxidation states, coordination numbers, possible isomers of multi-ligand complex ions, contribution of the solvent molecules in the ligand sphere, etc.
The method how one can use frozen solution Mossbauer spetra to identify iron species in aqueous solutions will be demonstrated with special focus on iron chelates and their reactions with hydrogen peroxide resulting in the formation of peroxo adducts.

Session 4 - Reaction mechanisms and nuclear recoils, nuclear base spectroscopies, radiation geochronology, isotope effects / 253

INVITED LECTURE - Half lives of nuclides for geological use: 2012 evaluations for ^{87}Rb, ^{235}U and ^{234}U

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The IUPAC-IUGS joint Task Group "Isotopes in Gesciences", TGIG, has evaluated the published measurement results for the decay constant (i.e. half life) of 87Rb and updated those of 235U and 234U relative to that of 238U. A significant part of our evaluation was the effort to follow strict metrological criteria (VIM, 2012) in our assessment of the measurement uncertainties according to GUM (2008).
The 87Rb half life was estimated by two groups using totally independent approaches. Nebel et al. (2011) compared Rb-Sr and U-Pb ages of different minerals, whose geological context suggests contemporaneity. This approach, which also applies to the comparison between 238U-206Pb and 235U-207Pb ages mentioned below, assumes that certain natural samples behave "ideally", i.e. all the relevant ages are expected a priori to be equal. This assumption is only as good as the independent control on the samples' petrology, and it is possible to underestimate systematic bias. Rotenberg et al. (2012) measured the radiogenic 87Sr accumulated in a batch of Sr-free Rb salt after 35 years. This approach relies on having performed precise and accurate measurements of the concentration and isotopic composition of the Sr present in the $RbClO_4$ at the time of crystallisation. The two sets of experiments yield indistin¬guishable results, which is a good indication that systematic biases were either coincidentally of the same magnitude and direction in two radically different experimental designs, or negligible altogether. The resulting best estimates are $\lambda 87 = (1.395 \pm 0.002) \times 10\text{-}11$ a-1 (1s uncertainty), $t1/2 = 49.7 \pm 0.1$ Ga.
The half life of 235U, which we had presented last year at the INCC congress (Villa et al., 2011, and references therein), has been slightly revised. It was brought to the attention of the Task Group that some workers reported a mass-independent fractionation of incompletely understood origin that affects odd- and even-mass isotopes in a different way (e.g. Amelin et al., 2005). Moreover, recent reports on N(238U)/N(235U) number ratio measurements in the same zircon samples used for geochronology (Hiess et al., 2012) indicate an individually variable number ratio, on average lower by (0.031 ± 0.011) % relative to the previously assumed value of 137.88. Including both effects into the 235U half life re-evaluation gives $t1/2 = 703.41 \pm 0.19$) Ma (1s uncertainty), $\lambda 235 = 0.98540 \pm 0.00027$ Ga-1.
These two effects do not modify the 234U half life relative to our 2011 assessment: $t1/2 = 245.44 \pm 0.16$ ka (1s uncertainty), corresponding to $\lambda 234 = 2.8241 \pm 0.0018$ Ma-1.

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INVITED LECTURE - Rapid Radiochemical Analysis of Radionuclides Difficult to Measure in Environmental and Waste Samples

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With the increasing requirement on rapid reaction system fot nuclear emergency preparedness, increasing numbers of nuclear facilities being decommissioned, as well as radioecological investigation, a large number of environmental, biological and waste samples need to be rapid analyzed for various radionuclides. Except gamma emitting radionuclides, other radionculides have to be first separated from the sample matrix and purified from the interferences before measurement, therefore defined as radionuclides difficult to measure. The traditional analytical methods for these radionuclides normally take a few days to weeks for one analysis; this makes the emergency reaction impossible in one day, and impossible to analyse a large number of sample from decommissioning work and radioecological investigation. In recent years, considerable efforts have been focused on the development of automated and rapid analytical methods. In our laboratory, automated analytical systems have been established by exploiting flow/sequential injection approach, using chromatographic separation concept, combined with rapid measurement of radionculides using ICP-MS. A number of analytical methods have been developed to determine 99Tc, 237Np, and isotopes of Pu and U in biological, environmental and nuclear waste samples. This work aims to summarize these automated and rapid analytical methods for the determination of various radionculides.

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INVITED LECTURE - TRIGA-SPEC: an apparatus for high-precision mass spectrometry and laser spectroscopy on short-lived neutron-rich radionuclides produced at the research reactor TRIGA Mainz

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High-precision measurements of ground-state properties of exotic nuclides such as nuclear binding energies, spins, radii, and moments, can be used to test nuclear models far away from the valley of stability. TRIGA-SPEC, located at the research reactor TRIGA Mainz, aims to investigate the ground-state properties of short-lived fission products produced by thermal-neutron-induced fission of U-235, Pu-239, or Cf-249. These fission products are extracted from the production site with a gas-jet transport system into an ion source, where an ion beam of the radioactive nuclides is formed. Subsequently, the ions are mass separated and prepared in a radiofrequency quadrupole cooler and buncher for the experiments. The high-precision measurements are performed with the Penning-trap mass spectrometer TRIGA-TRAP and by collinear laser spectroscopy with TRIGA-LASER. These experiments serve also as a development platform for MATS and LaSpec at the FAIR facility at GSI. Recent results of the experiments in offline operation and the status of the online coupling to the TRIGA reactor for the radioactive ion beam production are reported.

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ORAL PRESENTATION - Local Fields at Nonmagnetic Probe Sites in a Perovskite La0.7Ca0.3MnO3

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Perovskite manganese oxides (AMnO3) are known to exhibit the effect of colossal magnetoresistance (CMR), a phenomenon that electrical resistivity undergoes a drastic change as large as five to six orders of magnitude by an applied magnetic field. Because of this unique physical property, much attention has been given to these oxides aiming at wide industrial applications. In order to realize practical use of these compounds, it is of importance to obtain information on local fields in the material as well as macroscopic quantities such as resistivity and magnetization.

From this point of view, we have applied the time-differential perturbed angular correlation (TDPAC) method to a study of a perovskite La0.7Ca0.3MnO3, which exhibits the CMR effect by the paramagnetic-ferromagnetic phase transition (Tc ~ 245 K). In our previous work, we successfully observed drastic change in the local field at the probe nucleus, 140Ce, between temperatures above and below Tc, which evidently suggests that the magnetic field produced by spins in Mn ions can be transferred to the probe at the A site. Since the 140Ce probe arising from the disintegration of 140La takes two possible oxidation states, 140Ce3+ and 140Ce4+, however, it remains unknown whether the observed magnetic field (= 6.9 (3) T at 240 K) is exclusively transferred from Mn spins or is produced in part by a 4f electron spin localized at the probe in the chemical state of 140Ce3+. In the present work, therefore, we employed nonmagnetic 111Cd(←111mCd) and 111Cd(←111In) as the probe nuclei to shed light on the local field without self-owned spins. In the session, the effect of the 4f electron is discussed.

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INVITED LECTURE - Update of GEN-IV reactors and lead cooled reactors

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The path to develop advanced nuclear reactors that are superior to current systems is described in the 2002 Roadmap Report entitled "A Technology Roadmap for Generation IV Nuclear Energy Systems" which was prepared by the Generation IV International Forum (GIF) in 2002. This roadmap defined challenging technology goals for advanced reactor systems in four major areas:
• Sustainability,
• Economics,
• Safety and reliability, and
• Proliferation resistance and physical protection.

Of the six systems identified as promising by the GIF roadmap, the three inherently fast reactors are the GFR, SFR and LFR. Fast reactor systems, offer the potential for great efficiency in fuel utilization, and can be considered a superior option to achieve the goal of long-term sustainability.

The Fukushima accidents have drawn increased attention to the need for advanced reactors to be resilient in the face of unforeseen and/or severe accident conditions. In this context, the intrinsic characteristics of lead as a coolant lead to particular advantages of the LFR as an advanced reactor technology to meet present and future needs.

An extensive R program related to heavy-metal cooled systems was recently initiated in Europe. These efforts, conducted under the of EURATOM projects of the 6th and 7th Framework Programme, are addressing many of the most important issues related to the viability of the LFR.

In the ELSY project of the 6th Framework Programme, effort has been spend to improve compactness for economics and to improve the plant performance with respect to seismic loads. The abovementioned goals require large innovation of the primary system configuration and the use of innovative components which need extensive experimental campaigns devoted to qualification and functional confirmation.

In Europe the Romanian Government has expressed interest to Euratom in hosting the Demo of the LFR. Russia maintains interest in LFR and recently Chinese Organizations have started involving in LFR technology.

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INVITED LECTURE - Nuclear energy chemistry and recent progresses in nuclear fuel reprocessing in China

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Nuclear energy chemistry is one of the frontier areas of chemistry with high impact on national security, energy supply, scientific advances, social and economic development. Nuclear energy chemistry in China is now experiencing a renaissance, which is being strongly motivated by China's huge demand for nuclear energy. In this presentation, the progress in nuclear energy chemistry of China is selectively addressed. Some hot topics have been summarized and the main research results achieved by Chinese scientists in this field are highlighted, with emphasis on the chemistry of nuclear fuel cycle, such as front-end chemistry, materials chemistry and nuclear fuel fabrication, actinide chemistry and nuclear fuel reprocessing as well as nuclear waste disposal. Some measures about how to promote the radiochemical education and research in China are suggested, and future perspectives are briefly outlined as well.

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INVITED LECTURE - Nuclear Fuel Cycle: Processes, critical aspects and perspectives

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The growing awareness of the urgent need to reduce emissions of greenhouse gases and to increase the security of energy supply are changing significantly the energy scenarios of the world and low-carbon energy technologies in the next future will play a crucial role. To this end, although severe accidents occurred over the years, many countries are still considering nuclear energy as an important source for power generation - economically, environmentally and socially sustainable. Current technologies and the latest generation of nuclear plants offer better performance and reliability for all 60 years of their useful life (and beyond), increased efficiency in the use of the fuel, and advanced safety systems, with very high availability factors (over 90%) and less production of radioactive waste.

However, some critical points for a rapid expansion of nuclear energy remain. The whole nuclear power generation and the related fuel cycle is a very complex process, which requires large initial investments and considerable costs for a long period of operation and decommissioning.

Uranium resources are limited. At the current burn rate with the present technology, the conventional resources of uranium will ensure supply only for some hundred of years and the possibility that a strong expansion of nuclear energy could limit the availability of uranium on the market at competitive prices is in itself a limiting factor.

The enrichment process is another critical point. A large expansion of nuclear energy requires more efficient technologies and further enrichment facilities. To lose the control of these installations increases the risk of illicit trafficking of nuclear materials and the proliferation of nuclear weapons, including terrorist threats. The international system of safeguards on nuclear technology and materials in general ensures an adequate level of security, but it must be strengthened for some aspects, avoiding the spread of sensitive knowledge and technology and reinforcing the international co-operation.

Another important issue is the radioactive waste produced at each stage of the fuel cycle, which together with the plant safety is of greatest concern to the public. The management and disposal of radioactive wastes is an essential part of the fuel cycle. Progress needs to be made in building and operating facilities for the temporary storage and disposal of spent fuel and high-level wastes in geological formations.

Moreover, since the irradiated fuel discharged from the present reactor fleet, entirely made of light-water reactors, contains significant amounts of potentially recyclable combustible materials - mainly plutonium (about 1%) and slightly enriched uranium (about 95%), while only about 3.5% are fission products and 0.1% actinides – large scale reprocessing plants to extract uranium and plutonium from spent fuel are in operation and for the future it is expected an increasing of the current capacity, mainly related to the next-generation nuclear systems.

They will be able to burn the most part of uranium, as well as plutonium and actinides, which are the most responsible for the longevity of radioactive waste. At this point the problem of long-lived waste is strongly reduced, minimizing the need for geological repositories and current reserves of uranium will last for millennia.

To this aim, several technologies are under development, but their full commercial deployment will be reached not before of some decades.

The present work presents and discusses some critical points of current fuel cycle technologies and processes, providing also an overview on the perspectives for an increased sustainability - efficiency, safety and reliability, economics, waste reduction, proliferation resistance, etc - of nuclear energy in the future.

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INVITED LECTURE - Solution reactors for production of Mo-99 and Sr-89 (via Kr-89)

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Molybdenum-99 is the most important and widely used medical radionuclide which production in required quantities is possible only by fission products of Uraniun-235. In clinical practice the share of Mo-99 usage reaches approximately 80% of the total amount of radioisotope diagnostic procedures in the world. Strontium-89 is used for oncology and anesthesia and is capable of replacing painkilling drugs. Innovative technologies for production of Mo-99 and Sr-89 using solution reactors have been developed in RRC "Kurchatov Institute". These technologies reduce the required reactor power by approximately 100 times, decrease amount of radioactive waste and are suitable for low-enriched uranium fuel in comparison with conventional production methods. The technologies have been experimentally proven and refined using 20-kW solution reactor "Argus" where the isotope samples of required quality have been produced. Mo-99 has been extracted from irradiated soluble nuclear fuel by pumping the solution though a sorption column. Accumulation of the isotope occurs during 5-day reactor operation at nominal power while the extraction is done after the reactor shutdown. Sr-89 is obtained from Kr-89 which is evaporated during reactor operation. The achieved results lead to further plans including development of Mo-99 and Sr-89 production line using 150-kW nuclear reactor.

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INVITED LECTURE - Conceptual design of a low power ADS with a 70 MeV proton beam for reasearch and training

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In the framework of research on generation IV reactors, it is very important to have the opportunity of using infrastructures specifically dedicated to the study of fundamental parameters in kinetics and/or dynamics of future, fast-neutron based, reactors, a capability not available for presently available zero-power prototypes.

We propose the conceptual design of an ADS with high safety standards, to be used as well as a training facility, but also good flexibility to allow for an wide range of measurements: safety is guaranteed by limiting both the power of the system to be less than 500 kW and the neutron multiplication coefficient to be around 0.95, by using plutonium-free fuel and diffusion by a solid lead matrix.
Lead has been chosen by considering this prototype to be a useful step towards the design of future LFR, as well as because it allows a harder neutron spectrum, to facilitating tests on actinides fission, as well

The system is intrinsically subcritical and it needs an external neutron source to be sustained.
Specific target of the conceptual design is to optimize design features of the core in such a way to meet previous requirements with the use of commercially available accelerator to reduce design costs and increase reliability.

The conceptual design considers 88 active elements made by a solid lead matrix of dimensions 92X92X1200 mm., each containing 81 uranium oxide fuel rods, enriched at 20% of U235. Protons, coming from a continuous cyclotron of 70 MeV in energy and 1 mA in beam current, are converted into neutrons by a beryllium target. Cooling is provided by helium gas, transparent to neutron and not subject to activation.

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INVITED LECTURE - High power superconducting proton accelerators for ADS and Gen-IV

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Accelerator Driven Systems (ADS) have been considered and deeply studied since mid nineties as a possible solution to transmute long lived minor actinides and fission products in order to reduce the radio-toxicity of nuclear waste repository. The ADS scheme has been one of the driving objective for the strong development since then of the design of high intensity proton accelerators with MW beam power. An accelerator design based on stringent availability and reliability requirements has been the consequence of the combination of a sub-critical reactor with a particle accelerator. While the ADS concept is not considered anymore as the principal solution for the nuclear waste problem, it is still part of the Gen-IV scenario and the high power superconducting linear accelerators are being continuously developed for spallation neutron sources, particle physics and material irradiation facilities for fusion reactor.

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INVITED LECTURE - Transmutation of minor actinides in the molten salt reactor recently studied in Russia

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Abstract

The extremely high solubility of PuF_3, AmF_3 and the fission product fluorides in the eutectics LiF-NaF-KF observed recently (Fig.1 and 2) [1,2] allows to create the efficient molten salt reactor – transmuter (MSRT) for transmutation of the minor actinides (MA) from the spent nuclear fuel [3]. This observation opens also the way to the development of the molten salt fast reactor (MSFR) [2] with U-Pu cycle and changes the general approach to its closed nuclear fuel cycle (CNFC) realization. The first results are presented, obtained by the wide collaboration of Russian Institutes in the framework of the Rosatom program "Strategy of the minor actinides transmutation in the closed nuclear fuel cycle" [1], particularly the main parameters of the efficient subcritical MSRT which can transmute ~300 kg Am/GW•year without Pu consummation in the equilibrium mode operation [3].

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2. A.M. Degtyarev and L.I. Ponomarev, "Fast molten salt reactor based on LiF-NaF-KF", Atomnaya Energia, 112, p.367-368, 2012.
3. A.M. Degtyarev, O.S. Feinberg, F.I. Karmanov, et. al., "Subcritical Molten Salt Reactor with fast/intermediate Spectrum for Minor Actinides Transmutation", Proc. GLOBAL-2011, Murahari, Japan, paper 386820, 2011.

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INVITED LECTURE - The very powerful UCN source at the reactor TRIGA Mainz - Application for precise measurements of the neutron half-life

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Ultracold neutrons (UCN) are free neutrons with very low kinetic energies corresponding to v~ 5m/s. They can be stored both in material and magnetic traps and thus observed hundreds of seconds limited only by their natural lifetime. The long observation times make it possible to perform precision experiments on the neutron's fundamental properties such as the search for its electric dipole moment. Recently a new powerful UCN source has been installed at the pulsable reactor TRIGA Mainz. This source is well adapted to perform storage type experiments even at small research reactors. We plan to use the UCN source for precision experiments of the neutron half-life using magnetic storage methods. A status report on the UCN source and its application will be presented.

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ORAL PRESENTATION - Adsorption of selected fission products on various forms of TiO_2 nanoparticles.

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Due to the high selectivity, radiation resistance, thermal and chemical stability, inorganic ion exchangers have being widely used in the treatment of reactor coolant and aqueous nuclear wastes. Among others, inorganic sorbents, hydrous titanium dioxide are proposed as the promising inorganic sorbent for the efficient separation of fission and corrosion products such as 137Cs, 90Sr, 239,240Pu, 60Co, 65Zn and 54Mn. Recently, new forms of nanometer sized TiO2 with unique ion exchange properties were obtained. The physicochemical properties of nanostructured titanates are highlighted and the relation between properties and applications are emphasized.

In the present work we synthesized and studied ion exchange properties of TiO2 in nanotubes, nanofibers, nanowires and nanoribbons forms. The TiO2 nanostructures were synthesized using hydrothermal procedure. Material obtained has the large specific surface and internal structure of grains. The size and the shape of obtained nanoparticles were characterized by SEM and TEM methods and specific surface by BET technique.

The measurements were performed on the titanium dioxide nanotubes (diameter >5 nm, length >100 nm) nanofibers (thickness >7 nm, length >10 μm), nanowires (diameter >5 nm, length >10 μm) and nanoribbons (thickness 5> nm, length >10 μm). All synthesized samples were examined for adsorption of 137Cs and 85Sr in 0.1 M NaNO3 and KNO3 solutions. Additionally, kinetics, the dependence of the sodium salt concentration and influence of pH were examined. All nanoparticles studied show high affinity for both investigated radionuclides.

The affinity of nanotubes for Cs+ and Sr2+ was slightly higher in case of nanotubes than other TiO2 nanoparticles. The obtained results indicate a wide range of applications TiO 2 nanostructures as adsorbents for the efficient separation of fission and corrosion products from nuclear wastes.

The work was supported by Governmental Strategic Project: Supporting technologies for the development of safe nuclear power nr SP/J/4/143321/11

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ORAL PRESENTATION - Development of Decontamination Method Using Ionic Liquid as a Medium for Treating Waste Contaminated with Uranium

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Most of the metal or bed material wastes generated from uranium enrichment facilities or uranium refining and conversion plants are contaminated by uranium fluoride compounds. It is desirable to recover as much uranium as possible from these wastes. Additionally, if these wastes are decontaminated up to a level regarded as nonradioactive waste, the decontaminated materials should be reused.

In this study, we have evaluated the feasibility of application of 1-butyl-3-methyl-imidazolium chloride (BMICl) and cholin chloride-urea (CCU) ionic liquids to the treatments of metal waste contaminated with uranium.
Dissolution experiments were carried out by adding UF_4 powder samples into BMICl and CCU under atmospheric conditions. UF_4 samples were completely dissolved in BMICl after around 6h at 100 °C, and 38% of samples were also dissolved in CCU after 5 h at 100 °C. Steel waste contaminated with UF_4 was also decontaminated using BMICl at 100 °C under atmospheric conditions. The uranium concentrations of metal waste were found to decrease below the clearance level (1Bq/g) within 3h.

Cyclic voltammograms of the sample solutions prepared by dissolving steel waste into BMICl were measured. The reversible and irreversible peaks were observed around 0 and -1.2 V, respectively. These peaks were assigned as one electron reduction process of Fe^{3+} and one electron reduction process of UO_2^{2+}, respectively. These results suggest that electrolytic deposition of uranium should be performed without mixing of iron deposit. Bulk electrolysis of sample solutions prepared by dissolving steel waste into BMICl was also carried out at -1.5V at 80 °C. The deposits were formed on a carbon electrode as cathode. In analyses of deposits using XPS, only uranium component was detected.

Consequently, we confirmed that BMICl and CCU are effective media for the decontamination of uranium waste and that uranium can be recovered selectively from BMICl solution containing iron by the electrolytic method.

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ORAL PRESENTATION - Selectivity of bis-triazinyl bipyridine ligands for americium(III) in Am/Eu separation by solvent extraction. Quantum mechanical study

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Selective separation of actinide elements from highly radioactive nuclear waste is the key issue for modern technologies of nuclear waste reprocessing. Partitioning of long-lived minor actinides, in particular americium, and their subsequent transmutation into short-lived radionuclides would lead to a significant reduction of long-term environmental hazard from this radiotoxic waste, and contribute to the development of safe nuclear power. Derivatives of 6,6'-bis([1,2,4]-triazin-3-yl)-2,2'-bipyridine (BTBP), tetra-N-dentate lipophilic ligands which selectively extract trivalent actinides (An) over lanthanide fission products (Ln) from nitric acid solutions to organic solvents (SANEX process), have been considered the most promising species for hydrometallurgical recovering (partitioning) the minor actinides from high-level radioactive waste.

Theoretical studies were carried out on two pairs of americium and europium complexes formed by the BTBP ligands, neutral [ML(NO3)3] and cationic [ML2]3+ where M = Am(III) or Eu(III), and L = 6,6'-bis-(5,6-diethyl-1,2,4-triazin-3-yl)-2,2'-bipyridine (C2-BTBP). Molecular structures of the complexes have been optimised and total energies of the complexes in various media calculated using the DFT method (Gaussian 09). Selectivity in solvent extraction separation of two metal ions is a co-operative function of contributions from all extractable metal complexes. The individual contributions depend on physico-chemical properties of the complexes and on their relative amounts in the system. Semi-quantitative analysis of BTBP selectivity in the Am/Eu separation process has been carried out, based on the contributions from the two pairs of Am(III) and Eu(III) complexes. To calculate the energy of Am/Eu separation, a model of extraction process was used, consisting of complex formation in water and transfer of the formed complex to the organic phase. The greater thermodynamic stability (in water) of the Am-BTBP complexes, as compared with the analogous Eu species, caused by greater covalencies of the Am–N than Eu–N bonds, is most likely the main reason of BTBP selectivity in the separation of the two metal ions. The other potential reason, i.e. differences in lipophilic properties of the analogous complexes of Am and Eu, is less important with regards to this selectivity.

Analysis of Am–BTBP and Eu–BTBP bonding in the complexes studied, performed with the use of theoretical methods: QTAIM, CMO and NBO, allows us to conclude that the origin of the selectivity of BTBP ligands for Am over Eu is due to different contributions into bonding from various atomic orbitals of the Am(III) and Eu(III) ions.

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ORAL PRESENTATION - Decomposition of boric acid solutions and evolution of gases under mixed thermal and fast neutrons and gamma radiation

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Usually radiolytic gases (such as oxygen and hydrogen molecules) are not detected in cooling system of a research reactor even under room-temperature irradiation conditions. However, the presence of certain amounts of boric acid, which is known as a water soluble thermal neutron absorber, produces an evolution of gases in significant quantity in the reactor at room temperature. To study the radiolysis of the cooling water, we irradiated several water samples containing natural, 10B-enriched, and mixed boric acid in the ranges of 0 to 2000 ppm for the function of 10B concentration.

The boric acid concentration increased the extent of water decomposition compared to the absence of boric acid because of the nuclear effect from 10B(n,α)7Li, which is due to the radiation issued from a 10B reaction with thermal neutrons. It is well known that the products of water radiolysis are molecular (H2, O2, H2O2) and radical (H, OH, e-aq, HO2) species, and that high linear energy transfer (LET) radiation of 10B(n,α)7Li produces more molecular species than radical ones. The high LET radiation of alpha rays, produced from the addition of boric acid in water, inhibits the recombination mechanisms of radicals and stops the chain reaction.

The radiolysis of a boric acid solution under mixed thermal and fast neutrons and gamma radiation was examined quantitatively and qualitatively using a inductively coupled plasma mass (ICP-MS) spectrometer for measuring the 10B and 11B concentration ratio; a flameless atomic absorption (AA) spectrometer for the concentration of a product, 7Li,; a gas mass (Gas-MS) spectrometer for the measurement of H2, O2, and H2O concentrations; and titration methods for the H2O2 concentration. The total amounts of produced gases will be discussed with a comparison of the theoretical calculation values.

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OPENING LECTURE - Recent advances in nuclear data research for medical radionuclide production

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Nuclear data play a key role in the optimisation of production routes of medical radionuclides. In general, the production data of all commonly used diagnostic and therapeutic radionuclides are well known. The international activities to standardise those data will be reviewed. Furthermore, some recent efforts to develop alternative routes of production of a few widely used radionuclides, such as 99mTc and 68Ga, will be briefly discussed.

Regarding research oriented radionuclides, great demand exists for novel positron emitters, e.g. to study slow biological processes and to quantify dose distribution in internal radiotherapy. Some recent studies related to the development of 64Cu (T½ = 12.7 h), 124I (T½ = 4.2 d), 86Y (T½ = 14.7 h), etc. will be described as typical examples. In general, the low-energy (p,n) reaction on highly enriched corresponding target isotope is successfully utilized. However, for production of several positron emitters, intermediate energy reactions are preferable. Another area of increasing interest is internal radiotherapy and the choice lies on low-range highly ionising radiation emitters, i.e. low-energy β-, α and Auger electron emitters. In recent years a large number of charged-particle induced reaction cross section measurements have been performed, especially to produce some important therapeutic radiolanthanides with higher specific activity than in reactor production, though only with partial success. Cross section measurements to develop some novel therapeutic radionuclides have been challenging, and interdisciplinary techniques were employed. This will be exemplified by studies on a few radionuclides, such as 67Cu (T½ = 2.6 d; Eβ- = 577 keV), 225Ac (T½ = 10.0 d; Eα = 5830 keV) and 193mPt (T½ = 4.3 d; Auger electrons).

The future perspectives of medical radionuclide production will be considered. The potential of use of high energy protons and heavier mass projectiles will be discussed.

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INVITED LECTURE - ^{68}Ge-^{68}Ga production revisited: new excitation functions, target preparation and separation chemistry

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The use of ^{68}Ga ($T_{1/2}$= 67.7 m, β^+ decay branching 89.1%) for clinical diagnostic PET imaging is increasing.
The fact that it can be obtained from a generator system with long lived ^{68}Ge ($T_{1/2}$ = 288 d) as parent and its versatile chemistry could make it the PET analogue of the ^{99}Mo-^{99m}Tc pair.
The preferred route to produce ^{68}Ge is intermediate energy proton bombardment of Ga-containing targets. The long half-life however means that long irradiations at high particle flux are needed to produce enough activity for a useful generator (typically 750 MBq). We developed high beam power withstanding targets using electro-deposition of Ga/Ni alloy (70/30%) on industrially used thick Cu backings covered by a 20 μm protective Au layer. Using AC Constant Current Electrolysis (current density 34 mA/cm^2) a uniform layer containing 51 mg/cm^2 Ga is deposited in 7 h from an acid bath containing Ga_2SO_4, $NiSO_4$ and H_2SO_4.
Using thin Ga/Ni targets produced with this technique (6 mg/cm^2 Ga) we measured cross sections for reactions, induced by protons (up to 65 MeV) and deuterons (up to 50 MeV), producing γ-emitting; radioisotopes with halflife longer than 2 h. With a stacked foil irradiation, followed by high resolution γ-spectrometry;, excitation functions for ^{68,69}Ge, ^{66,67,72}Ga, ^{69m,65}Zn were determined, relative to the monitor reaction ^{nat}Cu(p or d,x)^{62}Zn. The results are compared to the scarce literature values while from derived thick target yields possibilities to use the deuteron reaction are discussed.
A PC-controlled automated set up for delivery of nca ^{69}Ge is developed and implemented in an industrial system ready to be mounted in a hot cell.
The target layer is dissolved in a mixture of H_2SO_4 and H_2O_2 using a heated,flow trough stripper. Prior to the extraction into toluene, concentrated HCl is added (normality of the solution 0.4 M HCl). The back extraction of the nca ^{68}Ge is performed using very diluted HCl. As a last purification step, a cation exchange column is used to adsorb traces of Ga and Ni. Samples are collected for radionuclide and chemical purity analysis. The 40 ml bulk is passed trough a 0.22 μm filter for final filtration-sterilisation.
Overall chemistry yield, tested by ^{69}Ge tracer, is higher than 90 %.

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INVITED LECTURE - Radionuclide production studies by heavy ion beams

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`In last two decades our research group is involved in the heavy ion assisted production and separation of neutron deficient no-carrier-added radionuclides. The advantage of heavy ion assisted production is choice of wide range of projectiles and therefore increasing the possibilities of reaching desired radionuclide far from the stability line. The main disadvantage is low production cross section, which made heavy ion assisted radionuclide production unsuitable at present for clinical applications, though in some cases HI cross sections are comparable with ⍺-particle activation cross sections. The high current ECR ion sources may address this limitation in future. Nevertheless, the neutron deficient radionuclides are comparatively short-lived and therefore highly suitable for tracer studies including biological systems.
In the conference, I shall briefly discuss the recent results obtained by our group on heavy ion assisted production of radionuclides. This includes production of 149Tb, 149Gd, 73As, 75Se, 209-211At, 97Ru, etc. In many cases, we have measured cross section for production of these radionuclides and compared with the theoretical prediction. The efficient and simple separation chemistry has been developed in each case. Apart from conventional liquid liquid extraction techniques, we have also used aqueous biphasic extraction system (ABS), taking polyethylene glycol (PEG-4000) and solution of various salts for separation of no-carrier-added radionuclide from the target matrix. Due to the absence of organic solvents, ABS can be viewed as greener technique.

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Overview of PET radionuclide production methods.

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Positron Emission Tomography (PET) Imaging requires radiolabelled materials (biomarkers) incorporating radionuclides that decay by positron emission.
They are largely neutron deficient and are made with a charged particle accelerator usually a cyclotron.
The most important radionuclides are 18F, 11C, 15O and 13N although an increasing interest is being seen in the radio metals in particular 68Ga 64Cu and 89Zr.
In order to achieve useable production yields of any radionuclide certain criteria have to me met especially the nuclear reaction cross sections at the charged particle energies available with a typical PET cyclotron with a proton energy of 10 to 20 Mev.
Available charged particle beam current will determine the intensity of the charged particle irradiation and hence the number of useful nuclear reactions achieved.
However many other factors come into play in the practical use of cyclotrons and their associated targets to achieve radionuclide intermediates suitable for incorporation into PET biomarkers.
The two most important radionuclides used in the Medical applications of PET are fluorine-18 and carbon-11 and aspects of their effective production will be discussed in detail.
Reference will be made to the methods of production of the other less widely used PET radionuclides without significantly overlapping with other presentations at this meeting.

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INVITED LECTURE - Radionuclides and radiopharmaceuticals at POLATOM

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Radioisotope Centre POLATOM is a state owned laboratory in the structure of the National Centre for Nuclear Research. POLATOM develops techniques for practical application of radioisotopes in various sectors, among them majority of products and services used in health care. POLATOM is supporting domestic and international users in highly specialized radiopharmaceuticals and radiochemicals for nuclear medicine and related fields, among them a number of products utilizing radionuclides irradiated in the multifunctional MARIA Research Reactor. POLATOM's facilities are well equipped and certified for manufacturing of radiopharmaceuticals. We are also carrying out extensive research programs on the new developments in the application of radionuclides supported by national and international multidisciplinary collaborations.
The main research and development domains are:
1. Investigation of novel biomolecules as carriers for radionuclides and preliminary assessment of their diagnostic and/or therapeutic utility.
2. Development of technologies for the production of high specific activity radionuclides in nuclear reactors and accelerators, using highly enriched target materials and modern separation techniques.
3. Development of methods for radioactivity measurement and assessment of radionuclidic purity (determination of □, β, and □ impurities).
4. Chemical and pharmaceutical development as well as in vitro and in vivo assessment of biological activity of new radiopharmaceuticals for clinical applications.
A very good communication network with domestic nuclear medicine units and research institutes interested in radiopharmaceutical development as well as with several international research institutions has been developed, supported by scientific and applied grants. The Centre participates in research projects coordinated by the International Atomic Energy Agency (IAEA) as well as in the European cooperation program of scientific and technical cooperation COST.

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INVITED LECTURE - Reactor production of radionuclides for molecular imaging and targeted radiotherapy

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The University of Missouri Research Reactor Center (MURR) was instrumental in the development of two commercialized radiotherapeutic agents: Sm-153 [Quadramet®] for the palliation of pain due to metastatic bone cancer, and Y-90 labeled glass microspheres [TheraSphere®] for the treatment of liver cancer. MURR is now actively developing other radionuclides with potential for use as targeted radiotherapy of cancer. One approach focuses on attaching a radionuclide via a bifunctional chelator to a selective targeting biomolecule such as a peptide or an antibody. A second approach is the use of nanoparticles in which the nanoparticle is comprised of the radionuclide and allows for the delivery of multiple radionuclides per targeting moiety. Radionuclides for therapy have unique half-lives (dose rates) and beta energies (tissue penetration ranges) and in addition some have a low abundance of gamma emissions that allow for imaging and tracking dosimetry. Radiotherapy with the new targeting moieties requires a diverse library of radionuclides to meet their specific needs. For instance, radiolabeled peptides, antibodies and nanoparticles have shown promise for diagnosis and radiotherapy of cancer by targeting receptors over-expressed on tumor cells. Due to the low concentration of tumor-associated antigens, high specific activity radionuclides and/or novel nanoparticle complexing techniques are required. Current efforts at MURR have focused on developing high specific activity radioisotopes that can be attached to biomolecules, readily converted into nanoparticles and or incorporated onto nanoparticles and are taken up selectively by diseased tissues; they deliver radioactivity for diagnosis or treatment of disease selectively and minimize or spare damage to healthy or normal cells. These tumor targeting biomolecules can be radiolabeled with different radioisotopes tailored to treat different cancers and diverse patient needs. The reactor production of radionuclides and their incorporation into nanoparticles and biomolecules will be discussed in the context of their physical and chemical properties as related to their potential utility in medical research.

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INVITED LECTURE - Review of Mo-99 and other reactor radionuclide production in RIAR in terms of world demands

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Production of Mo-99 was established in Russia more than 20 years ago. First facilities were placed at the Institute of Physics and Power Engineering and Karpov Physico-Chemistry Research Institute (both in Obninsk city). They were focused on domestic supply and had limited production capacity. The IPPE facility was shut-down a few years ago. The Karpov Insititute' facility continues its operation.

Principal decision were made in late 2009 by Russian Government to establish a new production facility – modern, providing prospective needs of nuclear medicine actively developing in Russian Federation, and increasing export capabilities of Russian isotope producers as well. The concept of this new production plant is aimed in providing large-scale, reliable, sustainable and continuous supply of the product, which is fully complied with International Pharmacopeia requirements. The process should be realized on a contemporary equipment and control systems, comply with GMP requirements and be environmentally safe.

To provide continuous production two pool-type reactors (RBT-6 and RBT-10) are used for irradiation. Their current irradiation capacity allows for irradiation up to 4 targets/week, providing 2-2.2 kCi/target at EOB. The number of irradiation positions can be increased twice in a nearest future.

The facility for irradiated targets processing was constructed at Research Institute of Atomic Reactors site (Dimitrovgrad) during 2010 - mid 2012 jointly with Isotope Technology Dresden (ITD, Germany), that is a part of Gamma Service Group International, GmbH (Switzerland). The ITD Company has provided the ROMOL-99 process, and principal set of equipment for processing as well we radiation safety systems (Xe-trapping line, iodine filter tower). The first production line was designed and constructed by December, 2010, it was tested and improved during first half of 2011. The production was started in August, 2011, first batches of Mo-99 were supplied to Russian producer of 99Mo/99mTc-generators. During late 2011 – 2012 considerable improvements were made in the process operation mode, ensuring Mo-99 radiochemical yield appox. 80%, and product quality corresponding to European Pharmacopeia requirements.

The second production line was installed in a new building, constructed during 2011-2012. The facility consists of two independent sets of equipment (dissolvers, vessels for solution, systems for Mo-99 isolation and purification) thus providing increased production capacity of the entire facility and reliable operation of the plant. Some equipment units (container transport system, targets, wastes handling, vessels for temporary storage ets.) that do not effect the production capacity, are used jointly thus decreasing cost of the entire facility. The second facility was assembled late June, 2012, and currently it is on the commissioning. Regular full-scale production is scheduled for November-December, this year.

For more than 30 years RIAR routinely produces wide spectrum of reactor radionuclides. The Institute is a principal (the only in Eastern hemisphere) producer 252Cf and other transplutonium elements (Cm-244,248, Am-241,243, Bk-249). Production of these radionuclides is possible due to unique high-flux reactor SM operated by RIAR. They are supplied mainly as sealed sources, produced by original technologies. The resent achievement of RIAR is production of Cm-244 alpha-sources for APX spectrometer that was successfully landed on a Mars planet last August.

Availability of high-density neutron fluxes (>2.1015 cm-2s-1) allows for production of high-specific activity radionuclides, that is a principal feature of RIAR. They are Co-60 (>250 Ci/g), Se-75 (>1200 Ci/g), Ni-63 (up to 12 Ci/g), W-188 (up to 8 Ci/g), Lu-177 (>90 kCi/g), n.c.a. I-131, I-125, etc. In a number of cases fast flux BOR-60 reactor is used for irradiation, producing Gd-153 (>100 Ci/g), n.c.a. Sr-89 (>3000 Ci/g).

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ORAL PRESENTATION - A new route for polonium-210 production from a bismuth-209 target

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A new method is proposed for the production and purification of polonium-210 (Po-210). This method is based on the bombardment of a bismuth-209 (Bi-209) target with a 38 MeV alpha particle beam that conducts to the production of astatine-210 (8.1 hrs) which decays to Po-210. It is further purified from bismuth target by a wet method using a liquid –liquid extraction process with tributyl phosphate (TBP). The main challenge relies on the separation of traces of Po-210 from a solution containing macroscopic quantities of bismuth. Several parameters were studied (acidity, aqueous medium, extraction time, TBP dilution, organic solvent) for defining the best conditions. The retained purification process consists in four steps: (i) the dissolution of the Bi target in nitric acid 10M followed by a reconditioning in 7M HCl, (ii) the extraction of Po-210 in 90% TBP in para xylene (iii) washing of the organic layer with 7M HCl and (iv) the back extraction of Po-210 in 9M nitric acid. The optimised process leads to a solution of Po-210 with a global recovery yield of 93 % with good radionuclide and chemical purities. The obtained data allow as well proposing an extraction mechanism of Po-210 by TBP.

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OPENING LECTURE - The Road to Cyclotron Produced Tc-99m

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Researchers at the University of Alberta have demonstrated that it is possible to produce and extract clinically significant quantities of 99mTc via the 100Mo(p,2n)99mTc nuclear reaction. Cyclotron targets have been engineered which significantly enhance their power-handling capacity to allow for extended high current irradiation. In addition, a process has been developed which allows for the efficient dissolution of the cyclotron target substrate, separation of the 99mTc and unreacted 100Mo and recovery of the expensive 100Mo for recycling. The quality of the extracted 99mTc has been verified by appropriate quality control protocols and animal biodistribution studies were performed which culminated in the completion of the first human clinical trial comparing with cyclotron produced 99mTc with reactor produced 99Mo/99mTc. Details will be presented on each of the steps in this process that highlight our solutions to the major problems that were overcome to produce a reliable alternative source for this key medical radionuclide.
Acknowledgements: Our partners at the Centre Hospitalier Universitaire de Sherbrooke (CHUS) and Advanced Cyclotron Systems Inc. We also a gratefully acknowledge financial support from NSERC/CIHR (MIS 100934), the Non-reactor based Isotope Supply Contribution Program (NISP) of Natural Resources Canada, Alberta Cancer Foundation, Western Economic Diversification Canada and the Western Economic Partnership Agreement.

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INVITED LECTURE - Cyclotron production of radionuclides with medium-energy proton beams and high-power targetry

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At iThemba LABS, proton beams of 66 MeV are used for the routine production of radionuclides and for neutron therapy. Between treatment irradiations, the beam is switched to the radionuclide production vaults and vice versa. Dedicated beam time for radionuclide production is also scheduled at night.

In recent years, several facility upgrades and new additions had been geared towards increasing the radionuclide production capacity by increasing the beam intensity. The development of suitable targetry to exploit the higher beam intensities were pursued in parallel. In addition, a beam splitter was developed and put into routine operation, which makes it possible to bombard two radionuclide production targets simultaneously. These developments, together with the addition of a new vertical beam target station (VBTS) led to a three-fold increase in production capability.

The main accelerator developments to achieve higher beam intensities were the addition of flat-topping resonators to one of the solid-pole injector cyclotrons (SPC1) and the separated sector cyclotron (SSC) as well as an additional RF buncher in the transfer beam line between them. This also required improved beam diagnostics, including non-destructive beam position monitors for continuous display of the beam position as well as stray-beam monitors installed at regular intervals along all the high-energy beam lines leading to the radionuclide production vaults. Extraction of 66 MeV proton beams with intensities up to 300 micro-ampere is now routinely possible, which can be accommodated on certain targets if rapid beam sweeping is employed.

The evolution of the radionuclide production programme at iThemba LABS with emphasis on the developments over the last circa 5 years will be presented. This will include an account of our experiences with high-power targets, in particular the VBTS tandem targets for the production of Na-22, Ge-68 and Sr-82.

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INVITED LECTURE - Radionuclide Production at Accelerator with High Power Targets

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The isotope production facility at 160 MeV proton beam of linear accelerator in the Institute for Nuclear Research of Russian Academy of Sciences (Troitsk) has the following characteristics providing high power irradiation:
- 4□-cooling of the targets (from 1 to 14 targets at one time);
- slanting 26o-angle beam on the target;
- lithium beam window between accelerator vacuum and cooling water of the target cell;
- a system of 4 cooled graphite collimators with thermocouples monitoring the beam shape directly before the targets;
- beam sweeping providing heat distribution.
General requirements for target materials at high intensity accelerator beams are the following: high cross-section of the radionuclide in the particle energy range; high abundance of the main producing material; known and acceptable non-radioactive impurities; high temperature stability; high heat conductivity; high radiation stability; low vapor pressure; low interaction with target shell or cooling water; low toxicity of the main material and impurities in case of medical isotope production; acceptability for radiochemical processing. A compromise between these requirements is to be provided in the most cases. Calculation with ANSYS program ensures the correct choice of target material and irradiation regime.
High yields are resulted in producing 82Sr from metallic Rb-target in stainless steel shell, 117mSn from Sb-containing targets in graphite, Nb- or Mo-shells, 225Ac and 223Ra from Th-target in graphite, Nb- or Mo-shells, 22Na from Al-target, 103Pd from Ag-target, 72Se from GaAs-target, 68Ge from GaNi, etc.`
New approaches are considered to irradiate cycling liquid rubidium targets to produce large amounts of 82Sr, and cycling lead-bismuth targets for radionuclide production and on-line extraction.

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INVITED LECTURE - Recent advances in large scale isotope production at LANL

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Large scale production of isotopes proceeds at the Isotope Production Facility (IPF) of the Los Alamos National Laboratory (LANL), using intense 100 MeV proton beams. The main metric of this program's success is the reliable time-critical delivery of large quantities of high-purity medical isotopes, which in turn depends upon many aspects including robust high power targetry, availability of intense proton beams and the ability to modify the primary beam energy. The talk will highlight recent advances at LANL on these fronts, which will further elevate the institution's stature as a world leader in providing both large and small quantities of a wide variety of radioisotopes to users.

Successful efforts of the LANSCE accelerator team in improving the 201 MHz Drift Tube LINAC operations, resulted in recent demonstrations of a 40 MeV, 250 μA capability and of 100 MeV, 360 μA high current operation of the LINAC. These advances combined with the developing worldwide short supply of isotopes like Sr-82 and Ge-68 as well as the emerging demand for therapy isotopes such as Ac-225 intensified the LANL efforts to advance understanding of and control over failure mechanisms occurring in high power production targets. Advances include the development of a next generation of RbCl salt targets and Rb-metal targets for increased Sr-82 production, research focused on understanding of failure mechanisms of Nb encapsulated Ga targets used in the large scale production of Ge-68, and the development of a first generation high current Th metal target design for Ac-225 production.

Another exciting new development includes efforts focused on the parasitic utilization of secondary neutrons for smaller scale isotope production that will significantly expand the IPF production capability with no negative impact on the oversubscribed proton beam time.

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INVITED LECTURE - ARRONAX: on the way to the production of radio-isotopes with high-power targets

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ARRONAX, acronym for "Accelerator for Research in Radiochemistry and Oncology at Nantes Atlantique", is a high energy (70 MeV) and high intensity multi-particle cyclotron located in Nantes (France). It is designed to be able to extract two proton beams simultaneously (dual beam mode) up to 2*375µA. Beams can be delivered into 6 experimental vaults: Four are devoted to radionuclide production and equipped with an irradiation station connected to our hot cells via a pneumatic system; one contains a neutron activator developed in collaboration with the AAA company whereas the last one is used for basic research with low intensity beam (alpha radiolysis, radiobiology and physics).
ARRONAX main activity is non-conventional radioisotope production for nuclear medicine for both therapeutic use (67Cu, 47Sc and 211At) and imaging (82Sr/82Rb and 68Ge/68Ga generators and 64Cu, 44Sc).

As a first step, our work has been focused on the production of strontium-82, copper-64 and astatine-211. For that purpose, we have developed our own targets and modified the irradiation stations, purchased from IBA, to meet our requirements and lower the beam power density on target. With these changes, it is possible to irradiated deposit (Copper-64 or Astatitne-211) as well as encapsulated targets. These changes have allowed producing strontium-82 from RbCl target using 2*95 µA proton beams making ARRONAX one of the few places in the world able to produce this isotope for commercial use. These changes will also allow a scale up of our Copper-64 production. In the case of Astatine-211, an additional beam energy degrader has been designed and mounted on one of the beam line and is presently under testing.

In order to further increase the beam intensity on our targets, we plan to play with all the available parameters. In the case of Strontium-82, we are planning to change the target material in collaboration with INR Troitsk (Russia). Indeed, Rb metal is offering higher production yield and better thermal conductivity than RbCl. We may also try the wobbling of the beam. Finally we are thinking on a design of a new target station able to handle larger beam spot on target (we are currently limited to 20 mm in diameter).
All these changes will help us to take advantage of the high intensity available on our cyclotron.

The cyclotron ARRONAX is supported by the Regional Council of Pays de la Loire, local authorities, the French government and the European Union

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INVITED LECTURE - Use of radioactive targets for production of therapy radionuclides at the Brookhaven Linac Isotope Producer

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Our program is presently investigating the production of 67Cu, 86Y and 225Ac. The half life and beta emission of 67Cu have long been recognized as attractive for radioimmunotherapy. The short lived positron emitter 86Y should be useful for patient dosimetry measurement prior to high dose 90Y immunotherapy. The alpha emitter 225Ac may be attractive for treatment of micrometastases, but supply from its 229Th parent is extremely limited. The reaction routes we have chosen for these products are 68Zn(p,2p)67Cu at proton energy above 100 MeV, 86Sr(p,n)86Y at proton energy of 15 MeV, and 232Th(p, spall)225Ac at proton energy above 130 MeV, respectively. The 68Zn and 86Sr targets are expensive enriched isotopes for which recovery and reuse are needed for economical production. After the first irradiation these materials will contain longer lived radioactive 65Zn and 85Sr. Similarly 232Th is slightly radioactive to start. Standard target claddings at BLIP are sealed by electron beam welding, but the use of this method for radioactive material is not feasible. Therefore a target cladding that can be sealed remotely in a hot cell was required. The final capsule design comprises two aluminum disks with thin machined windows that are held together with 8 screws, situated on the periphery of the target and sealed with a silver coated stainless steel “C” ring. Calculations of thermal and mechanical properties that guided the capsule design will be presented, as well as initial experimental results.

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ORAL PRESENTATION - Thermochromatography study of volatile Tellurium species in various gas atmospheres.

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Lead-Bismuth Eutectic has been proposed as spallation neutron target and as a coolant for Accelerator-Driven System. One of the main issues of using LBE is related to the production of Polonium and its potential release.
Tellurium was chosen as surrogate in order to designing experimental set-ups for investigating gas-phase chemical properties of Polonium and its compounds.
Carrier-free amounts of elemental Tellurium, Tellurium oxides and hydroxides formed at 950 °C in various flowing carrier gasses were studied by thermochromatography in quartz glass columns.
Partitioning of the various compounds in quartz glass column was deduced measuring the □-emission from the long-lived 123mTe at 212 KeV.
Enthalpies of adsorption of Te, TeO, TeO2 and TeO3 on quartz surface were deduced from the observed deposition temperatures using a Monte Carlo method. The potential chemical reactions occurring at different temperature are discussed.

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ORAL PRESENTATION - Complexation of Cm(III) with 2,6-bis(5-(2,2-dimethylpropyl)-1H-pyrazol-3-yl)pyridine (C5-BPP) studied by time-resolved laser fluorescence spectroscopy

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Reducing the long-term radiotoxicity and heat load of spent nuclear fuel by separating long-lived radionuclides and converting them into shorter lived or stable nuclides by fast neutron induced fission reactions is the aim of the so called partitioning and transmutation strategy (P).[1] Key step is the separation of the trivalent actinides Am(III) and Cm(III) from the trivalent lanthanides in the SANEX process. Due to the great chemical similarity of these metal ions, an efficient separation is very challenging. Alkylated 2,6-bis(1,2,4-triazin-3-yl)pyridines (BTPs) belong to the most promising extracting agents.[2] Although a multitude of BTP-type ligands has been synthesized, the molecular reason for the high selectivity of these soft N-donor ligands is not fully understood. To gain a better understanding, further investigations on the complexation of An(III) and Ln(III) with BTP-type ligands with systematic variations in the aromatic system are of particular interest.

In the present work, the complexation of Cm(III) with 2,6-bis(5-(2,2-dimethylpropyl)-1H-pyrazol-3-yl)pyridine (C5-BPP) in methanol is investigated by TRLFS. Time-resolved laser fluorescence spectroscopy (TRLFS) is a very sensitive spectroscopic method which makes it possible to follow the complex formation of fluorescing ions like Cm(III) in solution.[3] Spectroscopic parameters like position and shape of an emission band provide information on the number and the type of coordinating ligands. For the Cm(III) complexation with C5-BPP, three different species, the 1:1-, 1:2- and 1:3-complex, with emission bands at 603.7 nm, 607.7 nm, and 611.6 nm, respectively, are found. The species distributions for various ligand concentrations are determined and stability constants are derived (log K03 = 14.8). As extraction from nitric acid solutions with the C5-BPP ligand is only possible in the presence of a lipophilic anion, the complexation of Cm(III) with C5-BPP and 2-bromohexanoic acid is investigated. It is found, that 2-bromohexanoate ions are coordinated to the metal ion at low C5-BPP concentrations and are successively replaced by the N-donor ligand in the inner coordination sphere.

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OPENING LECTURE - Nuclear and radioanalytical techniques in nanotoxicology research

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Currently, enormous progress is being made in producing a great number of nanomaterials. However, in spite of hundreds of nano-products produced and currently available on the market a huge health and safety questions remain unsolved, and the assessment of possible health risks of nanoscale materials before they become ubiquitous in every aspects of life is necessary. In order to reach this goal nanotoxicology research has become a new frontier for a scientifically-based assessment of the environmental and human health impact of nanomaterials and nanoparticles (NP). In this context, there are a large number of challenges for the analytical chemist involved in the different steps of nanotoxicology research such as the physico-chemical characterization of NP and their use in in vivo and in vitro experiments with laboratory animals and cellular moedels (behaviour in biological media, toxicokinetics, uptake in whole tissue/ cells, intracellular distribution, binding with biomolecules). However, nanotoxicology research has a strong multidisciplinary character and requires an integrated use of different analytical techniques such as spectrochemical, nuclear and radiochemical, specialized microscopy, bioanalytical, and molecular biology techniques.
The aim of this work is to highlight the potential role that nuclear and radiochemical techniques plays in mechanistically-based nano(eco)toxicology research carried out at ECSIN in collaboration with the radiochemical laboratory of the University of Milan-INFN Section.
Applications carried out are based on the use of radioactive metallic zerovalent or metal oxide NP (^{198}AuNPs, ^{110m}AgNPs, ^{60}CoNPs, ^{59}FeNP, ^{58}NiNPs and $^{57}Co^{59}Fe_2O_4$) as radiolabelled by neutron irradiation or via radiochemical synthesis by no-carrier-added (NCA) radiotracers.
The need of a new specialized figure of radiochemist, the radionanoanalytical chemist, will be also stressed.

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INVITED LECTURE - Radiochemical neutron activation analysis: the continuous need of this analysis mode

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Attempts are being made to replace radiochemical neutron activation analysis (RNAA) by other analytical techniques capable of low-level element determination, such as various modes of atomic absorption spectrometry (AAS) for single-element determination, various modes of mass spectrometry, especially inductively coupled plasma mass spectrometry (ICP-MS) for multielemental analysis and/or by accelerator mass spectrometry (AMS) for long-lived radionuclides. The reason is that that the use RNAA is associated with a higher work-load and radiation burden for personnel compared with non-destructive, instrumental neutron activation analysis (INAA), and sometimes also with the use of chemicals that are considered to be not environmental friendly (e.g., organic solvents). However, RNAA remains to be a method of choice, especially for low-level, low-uncertainty determination of selected elements, and in selected applications. The continuous need of RNAA has been accentuated by a recent recognition of neutron activation analysis (NAA) as primary method of measurement [1], disregarding whether INAA or RNAA is employed. It has been demonstrated that RNAA is the most powerful means of optimization of NAA in terms of achieving the lowest element detection limits and uncertainty of measurement [2,3]. Examples are presented of superior low-level determination of, e.g., silicon, vanadium, manganese, nickel, selenium, iodine, rhenium, mercury in biological materials and determination of REE in geochemical and cosmochemical samples by RNAA. The major application fields involve biomedicine, agriculture, and chemometry, namely certification of reference materials. Recent trends and achievements in RNAA are also briefly mentioned, e.g., a fast decomposition of biological materials by alkaline-oxidative fusion, new and/or amended separation methods, such as replacement of liquid-liquid extraction by solid phase extraction, use of nanoparticles, and use of “green” chemistry in separation of elements and their radionuclides.

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INVITED LECTURE - Nuclear analytical methods in prostate cancer diagnostics

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The contents of twelve trace elements in normal (n=37), benign hypertrophic (n=43) and cancerous tissues (n=60) of the human prostate gland were investigated by instrumental neutron activation (INAA) and by radionuclide-induced (109Cd) energy dispersive X-ray fluorescent (EDXRF) analysis. The contents of Ag, Co, Cr, Fe, Hg, Rb, Sb, Sc, Se, and Zn, and of Br, Fe, Rb, Sr, and Zn were measured by INAA and EDXRF, respectively. The content of Fe, Rb, and Zn in samples was determined by two methods and mean values of the element mass fractions were quoted as a result. Mean values (M ± SEM) for mass fraction (mg/kg, dry weight basis) in normal tissue were: Ag – 0.048±0.009, Br 40.6±5.6, Co – 0.045±0.004, Cr – 0.53±0.08, Fe – 115±6, Hg – 0.056±0.011, Rb – 14.4±0.9, Sb – 0.045±0.007, Sc – 0.029±0.005, Se – 0.70±0.04, Sr 2.5±0.4, and Zn – 1078±113. In our previous studies it was shown that Zn and Ca levels in prostate are almost one order of magnitude higher than in other soft tissues. The obtained means for Br and Cr mass fraction are more than two times higher than mean values of element content in skeletal muscle, liver and whole blood. So, the human prostate accumulates not only Zn and Ca, but also such trace-elements as Br and Cr. It was observed that in benign hypertrophic tissues the contents of Co, Cr, Hg, Sb, and Se were higher and the contents of Br, Fe, Rb, Sr, and Zn were equal to those in normal tissues. The contents of Co, Rb, Sc, and Zn were lower and those of Ag, Br, Cr, Fe, Hg, Sb and Sr were higher in cancerous tissues than in normal tissues. Finally, we propose to use the in vitro and in vivo estimation of Zn mass fraction in gland as an accurate tool to diagnose prostate cancer.

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INVITED LECTURE - The use of nuclear analytical techniques in the identification and investigation of metal- and metalloid-containing proteins

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Most metals and metalloids present in biological materials are bound to proteins where they have essential tasks as part of the catalytic centers of enzymes or as structural components. It has been estimated that in the biosphere a multitude of these compounds exists but so far relatively few of them have been detected. Since in most cases the presence of a metal or metalloid in a protein cannot be recognized from its genetic code, specific methods for the identification of novel metal- and metallo-containing proteins and the investigation of their characteristics have to be applied. The information obtained in this way helps elucidate the biological functions of trace elements and is thus of great interest in many fields of the life sciences.

As the properties of most of these compounds are not yet known, initial information on their presence in tissues or cells can only be obtained by protein separation and determination of the element contents in the isolated fractions. For these investigations gel electrophoretic separation procedures have been combined with radiotracer techniques and nuclear analytical methods which allow surface scanning such as proton-induced X-ray emission or synchrotron radiation X-ray fluorescence. In the analysis of several trace elements in purified proteins or protein subunits, which are usually isolated in only very small amounts, neutron activation analysis has been successfully applied. The identification of a novel metalloprotein is then achieved by analysis of the amino acid sequence of the purified protein using MALDI-MS of the peptide fragments, mRNA determination and cDNA synthesis. In this overview the applications of nuclear analytical methods in this field of research and their advantages and disadvantages are discussed with the help of some examples.

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INVITED LECTURE - Exploitation of accelerator waste for the production of exotic radionuclides

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High-energetic protons and secondary particles produce in interaction with matter – due to the broadness of the induced nuclear reactions – a big variety of radionuclides, with some of them being very rare, exotic, and, in several cases, difficult to be produced by complementary reactions. Depending on the nature of the activated material, valuable isotopes, interesting for scientific and technological applications, can be extracted from samples stemming from the surroundings or components of a particle accelerator, especially if the initial proton flux is comparable high (in the Megawatt range).
Prominent examples are 7Be, 60Fe, 44Ti, 26Al, 53Mn and others, which play an essential role for our understanding of the synthesis of elements in different stages of the stellar evolution. Suitable sample materials for basic studies of the key nuclear reactions as well precise determination of the corresponding half-lives are therefore urgently needed.
Since PSI operates the most powerful high-energetic proton accelerator world-wide, this facility is best-suited for a R program aimed to "mine" such isotopes. An initiative called ERAWAST (Exotic Radionuclides from Accelerator Waste for Science and Technology) was started in 2006 with an ESF-funded workshop in order to identify and motivate potential users. After five years, in the frame of a second workshop, first achievements as well as realistic future plans for front-end experiments were presented.
The present contribution summarizes the most prominent results and gives an outlook on the upcoming experiments in the frame of the ERAWAST program.

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INVITED LECTURE - Novel mesoporous materials for actinide and lanthanide separation

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We have undertaken the design, synthesis, and testing of reusable mesoporous materials for actinide and lanthanide separation, sequestration, and sensing. An experimental-computational collaboration has yielded several materials with a high binding capacity for 239Pu: functionalized mesoporous silica, mesoporous carbon, and ferrihydrite. Advances have been made with all three types of materials. The affinity for target actinides of several ligands grafted to mesoporous silica, as well as their binding geometry, has been predicted by ab initio relativistic density functional theory (DFT) geometry optimization calculations, and compared to experimental batch sorption and X-ray absorption spectroscopy (XAS) measurements. Additionally, these batch studies have established the ability to reuse the materials as high-capacity sorbents. Ordered mesoporous carbon (OMC) has proven far superior to activated carbon in terms of Pu(VI) capacity and sorption kinetics. Batch and XAS studies of mesoporous and nanoparticulate ferrihydrite interactions with Pu(VI) have revealed interesting light-induced redox chemistry at the iron-oxide surface that may yield separation capabilities. Alternative forms of these materials, such as monoliths and thin films, are being explored but functionalization is a major challenge. We are expanding our studies to different actinide and lanthanides ions and examine the effects of the presence of competing ions, as well as systematic ligand variation. We are exploring OMC-based electrodes offer a promising route for electrochemical separation and sensing of actinides and lanthanides.

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ORAL PRESENTATION - Radiolabelling of engineered nanoparticles – different strategies for Ag^0-NP, TiO_2-NP and MWCNT

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Radioactive tracers provide a simple and effective tool for transport studies of nanoparticulate materials within environmental samples at laboratory scale. Compared to classical approaches radiolabelling of nanoparticles (NP) offers advantages in sensitivity and selectivity together with the possibility of in-situ imaging of transport phenomena. Particularly with regard to quantitative evaluation for transport studies radiotracers allow an easy differentiation between the elemental/nanoparticulate background concentration and the NP-derived input into an environmental sample. The limiting factor of the use of radiotracers is the possible alteration of experimentally relevant physical/chemical properties of the NP due to the radiolabelling and also the stable binding of the tracer on the NP. Depending on the experimental needs (half-life, decay-mode/radiation), different labelling methods are available.
The study aims at the comparison/evaluation of radiolabelling methods for Ag^0-NP, TiO_2-NP and MWCNT:

(1) direct activation of NP due to cyclotron irradiation:
- Ag^0-NP: $^{107}Ag(p,3n)^{105}Cd$ --> ^{105}Ag / $^{107}Ag(p,p2n)^{105}Ag$
- TiO_2-NP: $^{48}Ti(p,n)^{48}V$
- MWCNT: $^{12}C(p,3d)^{7}Be$

(2) self-diffusion of radioisotopes:
- Ag^0-NP: $^{105,110m}Ag$
- TiO_2-NP: $^{44,45}Ti$

(3) radioiodination
- MWCNT: $^{125,131}I$

(4) recoil labelling:
- Ag^0-NP: $^{7}Li(p,n)^{7}Be$
- TiO_2-NP: $^{7}Li(p,n)^{7}Be$
- MWCNT: $^{7}Li(p,n)^{7}Be$

The direct irradiation and the recoil labelling were carried out at a Scanditronix MC40 cyclotron [1, 2], self-diffusion experiments were carried by means of a Cyclone® 18/9 (IBA molecular) [3]. The methods were tested with respect to labelling yield, achievable activity concentration, pH-dependent stability of the labelling and the influence on NP-properties. Data thus obtained enable an appropriate selection of radiolabelling methods for different experimental applications.

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ORAL PRESENTATION - Synthesis of Silica-coated Bimetallic Nanoparticles as Radiotracers

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Bimetallic nanoparticles of Au with Ag, Cu, and Ir were synthesized by irradiating aqueous bimetallic ions with gamma radiation that generates hydrated electrons neutralizing the metal ions to a particle form. The bimetallic nanoparticles were coated with SiO2 making them chemically and physically stable when released into the environment and most of industrial processes for hydrodynamic investigation. Their shell and core diameters were recorded by TEM as 100~112 nm and 20~50 nm, respectively. The nanoparticles were then bombarded with neutrons in a research nuclear reactor in order to make them radioactive. The radioactive nanoparticles can play a role of radiotracers in refinery/petrochemical industries as well as in environmental resource hydrodynamic investigation.

Radioisotopes are useful due to their same chemical properties as those of stable isotopes and higher detection sensitivity than other types of tracers. Their usefulness has been demonstrated in various experiments in diverse areas including life science, medicine, and engineering. SiO2-coated radioisotope Au core-shell nanoparticles (Au@SiO2 NPs) were first synthesized by sol-gel reactions (the Stöber method) of aqueous Au NPs induced by irradiation without reduction agents. The physical integrity of the SiO2 coated nano shells in high gamma radiation dose environment was confirmed in the previous study.

This work reports the synthesis of radioactive silica-coated bimetallic NPs (Au-Ag@SiO2, Au-Co@SiO2, Au-Cu@SiO2, and Au-Ir@SiO2) by the Stöber method followed by neutron irradiation. The resulting particles can be used in natural resource investigations as activable tracers that are to be taken from the environment and activated in a nuclear facility for quantitative estimation of the tracer as a function of time and location.

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ORAL PRESENTATION - Nanomedicine Approaches of Radioactive Gold Nanoparticles In Cancer Therapy

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The most practiced approach of drug delivery in cancer therapy over the last century has involved intravenous administration of cytotoxic chemotherapeutic or radionuclide-based nuclear medicine agents. Limited affinity of targeted chemotherapeutic agents or radiopharmaceuticals to tumor cites and also the vascular and interstitial transport barriers continue to pose vexing challenges in achieving optimal therapeutic payloads at tumor sites. Therefore, effective therapy of solid tumors continues to be an unmet clinical need. Gold nanoparticles, because of their size, inherent therapeutic properties and targeting capabilities provide unprecedented opportunities for intratumoral delivery and are thus poised for major advances in cancer oncology. Nanoparticles derived from 198Au (a radioisotope of gold produced in nuclear reactors) are inherently therapeutic possessing ideal beta energy emission and half-life for effective destruction of tumor cells/tissue (βmax = 0.96 MeV; half-life of 2.7 days). We hypothesized that the intratumoral delivery of naturally available, non-toxic and FDA approved Gum Arabic functionalized radioactive gold nanoparticles (GA-AuNPs) will circumvent vascular and interstitial transport barriers resulting in targeted delivery of optimal therapeutic payloads with minimal/no toxicity to neighboring tissue. Detailed therapeutic efficacy studies in prostate tumor bearing SCID mice, injected with GA -198AuNP intratumorally, have demonstrated excellent retention of therapeutic payload (90%) and an overall tumor volume reduction of over 80% through a single intra tumor injection of GA-198AuNPs(1). These therapeutic efficay data provide compelling impetus for the clinical translation of this novel nanotherapeutic agent for treating a variety of solid tumors which include prostate, pancreatic and hepatic tumors in human patients. This presentation will present latest results on the utility of radioactive gold nanoparticles in prostate tumor therapy. This lecture will focus on presenting the realistic potential of GA-198AuNPs in the effective treatment of hormone refractory prostate and other solid tumors which normally manifest resistance in response to traditional cell killing therapies. New oncological approaches toward extending survival of prostate cancer patients as the GA-198AuNPs transform tumors from fast propagating to static stages will also be discussed.

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The LARAMED project at INFN Legnaro National labs

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In the framework of INFN SPES (Selective Production of Exotic nuclear Species) special project, aimed the new frontier of nuclear physics research of unstable nuclei, the creation of a center for innovative radionuclides production for radiopharmaceuticals is also pursued. Such a dedicated project, called LARAMED (Laboratory of RAdionuclides for MEDcine) will take advantage of the high performance SPES proton cyclotron (70 MeV, 750 microamps) which will be available at INFN LNL in the next coming years. The main goals of LARAMED project cover different topics, ranging from nuclear physics (excitation function experimental measurements), to engineering aspects (high power production targets) and radiochemistry issues (improvements of separation purification techniques). It will be a first step towards interdisciplinary researches directed to innovative radionuclides production, such as 64/67Cu, 82Sr/82Rb, 68Ge/68Ga which are of interest in nuclear medicine for diagnostic and therapy applications. Moreover studies and efforts are currently underway to start alternative accelerator-driven direct production of 99Mo/ 99mTc, (the most important and widely used radionuclide in nuclear medicine so far) according to pharmacopeia GMP rules, from 100Mo(p,xn) reactions. The goal is to get enough production to fulfill the hospital needs in north-east of Italy. Such researches will be a possible evolution of activities at Legnaro labs in the next years, which are a recognized excellence center in nuclear physics and nuclear astrophysics researches, towards a system where basic science and technological applications coexist and provide synergy to each other.

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Validation of neutron induced data up to 18 MeV for production of the therapeutic radionuclide 67Cu

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The radionuclide 67Cu can be produced via several routes. Initially the 67Zn(n,p)67Cu reaction was applied using a nuclear reactor. In a recent CRP of IAEA, the study of production of this radionuclide is in progress via the 67Zn(n,p)67Cu, 68Zn(p,2p)67Cu and 70Zn(p,α)67Cu reaction. In this work we have validated the neutron induced data for the production of 67Cu. The validation is based on integral test of the recommended data that were deduced from calculations done by using nuclear model codes STAPRE, EMPIRE, TALYS and ALICE-IPPE as well as by the statistical approach of fitting the experimental data.
Keywords: Radionuclides, Nuclear model calculations, Validation, Integral tests.

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Radiochemical separation of no-carrier-added 97Ru and 95Tc produced by 12C-induced reaction on natural yttrium target

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Due to favourable nuclear and chemical properties, the radionuclides, 97Ru; 2.83 d, -rays: 215.70 keV (85.62 %) and 324.49 keV (10.79 %) and 95Tc; 20.0 h, -rays: 765.789 keV (93.8 %), have been found promising for the investigation of delayed physico-chemical and biological processes. Various 97Ru/95Tc-labelled complexes have also been proposed for this purpose. Production of high purity radionuclides is therefore important. So far they have been produced either by light charged particle induced reactions or by neutron activation method. This paper illustrates simultaneous production of 97Ru and 95Tc in a heavy ion induced reaction and their subsequent separation from the bulk by ion exchange technique. A natural yttrium foil was irradiated by 75 MeV 12C6+ ions for 3.5 h. Thick target yields of 514 and 338 kBq/A-h were achieved for 97Ru and 95Tc respectively at the end of bombardment. The target was dissolved in 0.1 M HCl, spiked with 88Y (106.6 d), evaporated to dryness, and residue was taken into 0.01 M HCl. The nca 97Ru and 95Tc was then radiochemically separated from bulk yttrium by liquid-liquid extraction (LLX) using liquid anion exchanger trioctylamine (TOA) and liquid cation exchanger di-(2-ethylhexyl)phosphoric acid (HDEHP) dissolved in cyclohexane from HCl solution. More than 98% nca 95Tc was extracted into TOA phase possibly forming anionic species leaving nca 97Ru and yttrium in aqueous phase. Quantitative separation of 97Ru was achieved while bulk yttrium was extracted quantitatively into HDEHP (1-10%) from 0.1 M HCl. The technique offers efficient chemical separation of the products (97Ru and 95Tc) resulting very high separation factors (~106).

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A novel path in partitioning: Water-soluble BTP ligands for the innovative SANEX process

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With regard to an efficient partitioning process the separation of the trivalent minor actinides from the chemically similar fission lanthanides is one of the key issues. This separation process requires highly effective extracting agents. Alkylated bis(1,2,4-triazin-3-yl)pyridines (BTP) and alkylated bis([1,2,4]triazin-3-yl)-[2,2']bipyridines (BTBP) are among the most promising SANEX extracting agents, selectively extracting trivalent actinides from nitric acid solutions. Based on the conventional SANEX process an innovative SANEX (i-SANEX) process has been developed, combining the SANEX with the previous DIAMEX step. Hydrophilic aq-BTP is used as aqueous complexing agent for selective back extraction of actinides from organic phases loaded with trivalent actinides and lanthanides. Our studies focus on the complexation of trivalent actinides and lanthanides with aq-BTP (a) to gain fundamental knowledge on the unique properties of BTP, (b) to compare BTP with aq-BTP, and (c) to determine complex formation constants for trivalent actinides and lanthanides required for future process development. In the present work complexation studies are performed with Cm(III)/Eu(III) using time-resolved laser fluorescence spectroscopy (TRLFS). With increasing ligand concentration the following complexes are identified and spectroscopically characterised: [M(aq-BTP)], [M(aq-BTP)2], and [M(aq-BTP)3] (M: Cm(III), Eu(III)). From our spectroscopic results the stability constants and thermodynamic data of the stepwise complex formation are derived. A comparison of the log K values of the [M(aq-BTP)3]-complexes (M = Eu(III), Cm(III)) shows a significantly higher stability constant for trivalent actinides, which is in perfect agreement with the selectivity observed in liquid-liquid extraction tests.

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Pd-based intermetallic targets for high intensity irradiations

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The stability of actinide targets during the irradiation with intense heavy ion beams is a prerequisite to successful experiments with super heavy elements [1]. Due to higher thermal conductivity, electrical conductivity and mechanical stability metallic targets are suggested to be superior to the widely used nowadays solely electroplated ones. Recently we proposed a simple method which allows producing Pd-based intermetallic targets for high intensity irradiations [2]. Based on the molecular plating technique [3] followed by coupled reduction [4] this method was successfully applied to different lanthanide and actinide isotopes. 3 μm Pd backing foils were found to be the most suitable for preparing pinhole-free targets [5].
First irradiation experiments with intermetallic targets were carried out in November 2011 at the Oslo Cyclotron Laboratory, University of Oslo, Norway. 0.73 mg/cm2 238U/Pd target was irradiated at the MC-35 Scanditronix cyclotron using a 0.5 μA proton beam with cyclotron energy of 30 MeV.
Two 243Am/Pd intermetallic targets (0.85 and 1.7 mg/cm2) have been prepared and irradiated at the U-400 cyclotron at the Flerov Laboratory of Nuclear Reactions in March 2012 for several days with intense beams of 48Ca. Both 243Am targets were characterized by alpha-particle spectroscopy and light microscopy before and after irradiation. For direct comparison, the performance of a 'classical' electroplated 243AmO2/Ti target was examined.

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Development of actinide liquid scintillating target

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We present the development of a new kind of active actinide target, based on organic liquid scintillators containing the dissolved isotope that can be used for fission studies, but also for (n,xn) reactions measurements and potentially for radiative capture experiments. Amongst many advantages one can mention the very high detection efficiency, the Pulse Shape Discrimination capability, the fast response allowing high count rates and good time resolution and the ease of fabrication. The response of this target to fission fragments has been studied because the response of organic liquid scintillators to fission fragments is almost unknown. In the present work the response to fission events was also obtained with a 252Cf source. Several kinds of scintillators were used. Especially it was shown that a scintillator based on Di Isopropyl Naphthalene (DIN) solvent with the PBBO scintillating molecule lead to a very good separation between fission and proton recoils The light quenching against the actinide dissolved mass has been characterized. For uranium even if a strong quenching is observed, concentrations up to 10 mg/mL can be envisaged depending on the experiment to be performed and on the geometry of the active target. For thorium no quenching was observed. The discrimination of alpha, fission and proton recoil events is demonstrated. The alpha decay and fission detection efficiencies are simulated and compared to measurements. Finally the use of such a target in the context of fast neutron induced reactions is discussed.

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Preparation of TBq Activity 7Be from SINQ Cooling Water

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7Be is an important radionuclide for investigations of several astrophysical processes and phenomena. The study of the destruction of 7Be during the first 10-15 minutes of Big Bang Nucleosynthesis (BBN) via the 7Be(n,alpha)alpha reaction could help to understand the longstanding problem in BBN theory - the disagreement of the predicted abundance of primordial 7Li with the observed one. Another application of 7Be is the study of key reactions concerning the solar neutrino flux, in particular the reaction 7Be(p, γ)8B.
10Be and its daughter products have been used to examine soil erosion, soil formation from regolith, as well as variations in solar activity and the age of ice cores. One of the "hot topics" is the half-life of 10Be, where the literature values differ from 1.34 to 1.51 My. Two very recent measurements support rather the lower value: 1.388 My and 1.386 My [1, 2]. Additional measurements are, therefore, urgently needed. One possibility is the use of LSC for the determination of the activity and ICP-MS for measuring the number of atoms. The mass-bias calibration of the ICP-MS requires at least 2 mass points in known amounts and since Be has only one stable isotope (9Be), 7Be can serve as the second marker.
Another application area of this interesting and rare isotope is the development of new construction materials, requiring sensitive methods for studying their wear resistance. Implantation of 7Be and follow up the changes of its activity can determine wear-out less than a micrometer.
7Be is produced in considerable amounts in the cooling water (D2O) of the Spallation Induced Neutron Source (SINQ) facility at PSI by spallation reactions on 16O with the generated fast neutrons. By-products can be nearly neglected, so that this cooling water establishes an ideal source for highly active 7Be-samples.
A shielded ion-exchange filter containing 100 ml of the mixed-bed ion exchanger LEWATIT was installed as a by-pass for the cooling water into the cooling circle of SINQ for 6 weeks. The collected activity of 7Be was in the range of several hundreds GBq. Further the 7Be was separated and purified in a hot-cell installed, remote-controlled separation system. The facility is capable for production of 7Be with activities up to 1 TBq per year.

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Development of production possibilities of n.c.a radiomanganese in a non aggressive and toxic medium

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In view of new arising hybrid imaging methods, e.g. the PET/MRT combination [1], it is of high interest to develop new multimodal imaging tracers. In these imaging tracers rests the potential for the evaluation of old and new contrast agents in MRT, especially concerning new nanoparticle contrast agents. A large part of these newly composed MRT contrast agents are manganese based [2]. Therefore the cross sections for Cr-nat(p,xn)Mn-52g,m reactions and its byproducts (V-48,Cr-48,49,51) were measured up to 45 MeV, and a method was developed to separate n.c.a. radiomaganese from bulk chromium by ion chromatography.

Cross section measurements were conducted by the stacked-foil technique. Because of the brittle nature of metallic chromium an electrodeposition method [3] was used to obtain thin metallic layers on a gold backing. Multiple foil stacks were irradiated with protons of 17 or 45 MeV incident energy. The measured cross sections differ only slightly (1-10%) from literature, except for the Cr-nat(p,xn)Cr-51 reaction which shows a massive divergence (≈400%) from published data [4]. This ascertained our previous measurements [5].

For the development of the separation process cast plates of chromium were irradiated with protons in the energy range of 17 to 7 MeV. These targets were dissolved in fuming HCl. Bulk chromium was then separated from n.c.a. Mn-52 by means of ion chromatography with the cationic exchange resin DOWEX 50WX8. A quantitative separation was achieved with 0.1 M sulfuric acid as eluent for chromium and a 0.067 M ammonia citrate solution with pH 7.3-7.4 for n.c.a. radiomanganese. The latter eluent is not aggressive and not toxic. The separation took 3-4 h and 99% of Mn-52 was eluted from the column in ca. 12 ml of the eluent.

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Separation of carrier free 177Lu from 177Lu /Yb mixture by electro-amalgamation of ytterbium

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The process of isolation of no-carrier added 177Lu produced via 176Yb(n,gamma)177Yb - 177Lu from mixture containing macroscopic amounts of the ytterbium target material was investigated. For this purpose a novel method of electrochemical selective amalgamation of ytterbium from 177Lu/Yb mixture into mercury-pool cathode was applied. The electrolyte solution contained mixture of 20 mg ytterbium in 5M HCl and 177Lu as 177LuCl3 in 0.15 M sodium citrate. As anode platinum plate was used. In order to develop an optimal condition of amalgamation of ytterbium, effects of pH of the electrolyte solution, potential and time of the electrochemical process as well as number of cycle of electrolysis performed under the same conditions using fresh electrodes were examined. The concentration of lutetium and ytterbium in the electrolyte solution was determined by spectrometry ICP-OES. The best performance of the method, allowing cementation up to 94 % of ytterbium from the 177Lu/Yb mixture was obtained at pH of 6-7, potential of 8 V, time of 60 min and two cycles of the electrolysis. Concentration of 177Lu in the solution after fixing ytterbium in amalgam remained at the same level. As results of such defines process molar ratio of Yb:Lu was reduced from 3000 to 170. The atomic absorption spectroscopy (AAS) measurement showed that the content of mercury in the solution containing 177Lu after cementation of the ytterbium was bellow detection and determination limit level amounted respectively 0.3 and 0.6 µg/kg.

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Preparation of 57Co sources for Mössbauer Spectroscopy

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The method of preparation of 57Co source applied in Mössbauer Spectroscopy was developed. This method comprised electrodeposition of carrier-free 57Co on rhodium foil followed by thermal diffusion of 57Co into rhodium matrix. A series of experiments were performed in order to determine the optimal conditions for electrodeposition of cobalt on rhodium foils 6um thick. Electrochemical cell consisting of platinum anode and rhodium disc as the cathode was chosen. The electrolyte was an aqueous solution of ammonia citrate 25g/l, hydrazine hydrate 25/l and carrier-free 57Co in the form of 57Co(II) in 0,1M HCl. The diffusion of 57Co into rhodium lattice was carried out by annealing the foil at temperature of 1100°C at high vacuum, in quartz tube. The 57Co active cores were encapsulated in cylindral Ti capsules with Be windows. The Mössbauer spectra were measured to verify the quality and efficiency of the testing sources. The experiments performed allow making an optimum choice of the electrodeposition parameters of carrier-free 57Co on rhodium foil. The highest efficiency approaching 100% and the best rate of deposition were obtained using current density 50mA/cm2 and electrolyte volume – 5ml. The best results of diffusion of electrodeposition cobalt-57 onto rhodium matrix was obtained in an annealing process at 1100°C in vacuum over 10-6 hPa. The main spectra parameters of the prepared sources are fairly acceptable with respect to the typical obtainable values for α-Fe absorbers in Mössbauer spectroscopy. The results obtained confirm that the deposited layer diffused almost completely into rhodium matrix without substantial loss of the activity deposited.

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Accelerator-based Alternative Tc-99m production: EMPIRE 3.1 theoretical simulations of cross sections for Mo(p,X) reactions and comparison with literature experimental data

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Technetium 99 metastable is the most important and widely used radionuclide in nuclear medicine for over 80% of all diagnostic procedures. Recent shortage of this isotope prompted the international scientific community to ask for investigations on new possible production routes. As possible alternatives to the current reactor-based $^{235}U(n,f)^{99}Mo \rightarrow ^{99m}Tc$ scheme, accelerator-based methods have been considered. In this work, a feasibility study for the production of ^{99m}Tc from irradiation with protons on natural and highly enriched molybdenum targets has been performed. In the framework of INFN LARAMED (LAboratory of RAdionuclide for MEDicine) project currently under way at Legnaro labs, theoretical simulations of excitation functions and yields have, in particular been studied for $^{nat}Mo(p,X)$and $^{100}Mo(p,X)$ reactions in the 5-35 MeV proton energy range. In addition, a review of all existing experimental data has been collected and compared with the simulations. Theoretical calculations were carried out using the new EMPIRE 3.1 (Rivoli Release) nuclear code, a modular system of nuclear models whose new features permit to evaluate the population probability of isomer states. Preliminary results of simulations have shown good agreement with experimental data. Estimation for the production of other radioactive and stable nuclides together with ^{99m}Tc has been calculated and considered an important discriminator for the feasibility of the production route since the presence of other harmful nuclides in the final product could affect the diagnostic outcome and radiation dosimetry in human studies.

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Cyclotron produced ^{45}Ti – production, purification and yields

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Studies of the environmental fate of nanoparticulate TiO_2 require suitable tools for tracing the nanoparticles in complex environments and media. A promising method is the isotopic radiolabelling of the TiO_2-nanoparticles with ^{44}Ti ($T_{1/2}$ = 47.3 a) or ^{45}Ti ($T_{1/2}$ = 3.08 h). Due to the different decay modes and half-lives, different experimental setups are accessible with these radioisotopes. The presented work is focused on the production and purification of the short –lived positron-emitting radionuclide ^{45}Ti (n.c.a.).
For this purpose, we used the nuclear reaction ^{45}Sc(p,n)^{45}Ti [1]. The irradiation was done at a COSTIS target station mounted at a 2 m beam transfer line of a Cyclone® 18/9 (IBA molecular). The mono-isotopic natural scandium allowed an easy target design. A scandium foil (thickness: 100 µm) was put together with an energy-degrading foil into a disk-like sample holder and was then transferred into the COSTIS target station. The irradiation was carried out with 12 MeV protons and a current of 20 µA for 20 min – 30 min.
Radionuclide separation and purification was done by means of ion exchange chromatography [2] or liquid-liquid extraction [3]. Both methods were compared. Higher yields and better purification results were obtained with liquid-liquid extraction. The yield of the liquid-liquid extraction was about 75 % - 80 %, n.c.a. ^{45}Ti stock solution (1 M HCl) had a activity concentration of up to 120 MBq/mL.

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Separation of no-carrier-added 109Cd from natural silver target using RTIL 1-butyl-3-methylimidazolium hexafluorophosphate

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The room temperature ionic liquid (RTIL), 1-butyl-3-methylimidazolium hexafluorophosphate [C4mim][PF6] has found application in separation of a range of metal ions replacing volatile and toxic traditional organic solvents in liquid-liquid extraction (LLX) systems. Despite of disadvantage of probable release of HF in acidic reaction, the RTIL [C4mim][PF6] is used widely in developing green processes in analytical chemistry as it is hydrophobic in nature, lacks vapor pressure, and can be synthesized easily. Favorable half-life of 109Cd (461.4 d) allows its use in the field of medical science, environmental science as well as in technology. Several reports are also available on the production and separation of no-carrier-added (NCA) 109Cd. This paper reports the separation of NCA 109Cd from the natAg target using [C4mim][PF6] in LLX. The 109Cd was produced by bombarding a natAg foil (25.4 mg/cm2 thick) by 30 MeV α-particles at the Variable Energy Cyclotron Centre, Kolkata, India. After the decay of all short-lived products, NCA 109Cd was separated from the bulk Ag using [C4mim][PF6] as extractant in combination with HNO3 where ammonium pyrrolidine dithiocarbamate (APDC) was used as a complexing agent. At the optimum condition, 3 M HNO3, 0.2 mL 0.1 M APDC and 1.25 mL [C4mim][PF6], bulk Ag was extracted to the IL phase binding with APDC, leaving ~ 85% NCA 109Cd in the aqueous phase. The ionic liquid was also recovered by washing the IL phase with 5 M HNO3. The reported separation technique is simple, fast and in concurrence with green chemistry approach.

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Application of a mixed bed column for the removal of iodine from radioactive process effluents

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Safety and appropriate management of nuclear waste are of major importance in radioisotope production. Radioiodine has been identified as one of the most dangerous radioelements in terms of radiological effects on the population in case of accidental release; accordingly substantial efforts have been made to optimize the iodine capture in liquid effluents during the production process to avoid any possible iodine gas release from liquid waste during storage. The IRE production process results in highly acidic and oxidizing effluents, iodine might thus be present in various oxidation states and species. This needs to be taken into account for the decontamination of these effluents; accordingly a mixed bed column approach was chosen.

Literature reports the use of anion exchange resins and more recently of silver-coated sorbents such as zeolithes, alumina or activated carbon for iodine removal, the letter being very attractive as silver is known to interact very strongly with iodine. However, these sorbents show significant leaching of silver when used in acidic media and can thus not be used in the present case.

CL Resin (TrisKem International) is an extractionchromatographic resin that retains silver over a wide pH range including high acid concentrations. It was further shown, after being loaded with silver, to strongly retain iodine species forming insoluble complexes with silver making it well suited for use in the given context; XAD-4 Resin (Sigma) on the other hand is known to retain elemental iodine.

A mixed bed column based on silver loaded CL Resin and XAD-4 resin was developed and optimized for iodine removal from IRE's process effluents by varying column size and resin ratios. Prepared mixed bed columns were evaluated by treating multi-curies production process solutions at the IRE. The mixed bed columns showed high iodine removal from elevated effluent volumes (> 10 L) even at flow-rates up to 150 mL/min.

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64Cu and 67Cu Production and Purification Research at the Radiation Science and Engineering Center at the Pennsylvania State University

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The United States faces a shortage of medical isotopes for clinical use and for research and development of new therapeutic and diagnostic procedures. The Society of Nuclear Medicine and the National Cancer Institute in the USA have noted that 64Cu and 67Cu isotopes are in short supply. The isotope 67Cu (t1/2 = 61.83 h) emits a beta particle useful for cancer treatment and has several gamma-ray emissions appropriate for medical imaging. As a positron-emitter (17.9%) and a β--emitter (39.0%), 64Cu (t1/2 = 12.7 h) is used for positron emission tomography scans and to study copper behavior in the body for the diagnosis of metabolic diseases. The small amounts of 64Cu and 67Cu currently available are produced via charged particle reactions, but existing accelerators are in high demand for other applications. To expand the production capabilities for these two isotopes, the Radiation Science and Engineering Center at the Pennsylvania State University is investigating reactor-based production methods via the (n,p) reactions and . These reactions require neutrons with energies above 1 MeV, which are available in the central thimble of the Penn State Breazeale Nuclear Reactor at a flux of 1.6x1013 n/cm2/s. To reduce the production of unwanted radioisotopes, 99% isotopically enriched 64Zn and 94% enriched 67Zn are used. 64Zn and 68Zn can activate in measureable quantities to radioactive 65Zn (t1/2 = 243.8d) and 69Zn (t1/2 = 13.76 h) due to thermal neutrons; both types of targets can be shielded to reduce these activation products. Post-irradiation, quartz-encapsulated zinc oxide targets are dissolved in acid, followed by purification via one or more ion exchange methods. Although the cross-sections for these (n,p) reactions are low, our development of a reactor-based transmutation production method for 64Cu and 67Cu offers several advantages: large amounts of sample may be irradiated simultaneously and the product is carrier-free, making it highly isotopically and chemically pure. Experimental details and some results for this reactor-based alternative 64Cu and 67Cu production and purification method will be presented.

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Measurements of γ- and β+-intensities of Ti-45

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With rising interest in non-standard PET-nuclides also Ti-45 comes into focus. Its low maximum positron energy of 1.04 MeV in combination with the small amount of γ-rays (most intense at 720.22 keV: 0.15%) and its advantageous half-life of 184.8 min makes it a suitable nuclide, especially for studying some longer termed processes. There have been first investigations about its usability as nuclide in positron-emission-tomography studies [1,2]. However, the corresponding emission intensities have to be known very precisely for medical applications in order to minimize the radiation exposure of the patient and to enhance the diagnostic value by correct quantification.
Ti-45 was produced via the Sc-45(p,n)Ti-45 nuclear reaction. Metallic scandium served as target for irradiation at the Baby Cyclotron 1710 of FZ Jülich with protons of 12 MeV incident projectile energy. Copper foils as beam monitors assured the precision of the proton energy and beam current. After dissolving the irradiated target in 4M HCl, radionuclidically pure Ti-45 was isolated in n.c.a. form via ion chromatography, using 100-200 mesh Dowex 50WX8 resin and 4M HCl as eluent. The co-produced Sc-44 was used as indicator for the absence of scandium. Thin samples were prepared and investigated using positron-counting, x-ray spectrometry and γγ-coincidence measurements to determine the absolute positron intensity. The γ-ray intensities were ascertained relatively to the positron emission intensity using HPGe-detector γ-ray spectrometry.
The performed ⊠⊠- and ⊠⊠-coincidence counting resulted in positron emission probabilities of 82.4% and 84.9%, thus supporting the data given in the literature [3]. The γ-ray intensities were found to be smaller than those given earlier, showing deviations of up to 30% [4-6].

In this study reproducible decay data could be obtained which specify and improve the β+- and γ-intensity values reported so far, thus strengthening the reliability of these important data for quantification and corrections of PET-measurements.

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Production and purification of 56Co at the Leipzig cyclotron

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60Co (T1/2 = 5.27 a) is one of the most used radionuclide for sterilization of medical equipment, as a radiation source for medical radiotherapy, industrial radiography and food irradiation due to the high gamma-energy of 1.33 MeV. In case of release in the geosphere, e.g. soil and aquatic systems, the migration behaviour of cobalt is not well understood. For geochemical investigations, e.g. migration and adsorption studies in soil and rock formations, the short-lived isotope 56Co (T1/2 = 77 d) can be used.
We produced 56Co at a recently installed 18 MeV-cyclotron by using the nuclear reaction 56Fe(p,n)56Co.[1] The target was prepared by pressing metallic iron powder into an aluminium plate and cover it with an aluminium foil. After the irradiation with 11 MeV protons for 1 h at a current of 25 μA, the iron was dissolved with a mixture of concentrated HCl and concentrated H2O2.[2] The separation of 56Co from the target material was done by liquid-liquid extraction with methyl-tert-butylether (MTBE) from 5.2 M HCl.[3] Alternatively, an anionic exchange with DOWEX 1x8 as a column material can be used.[4] Due to the shorter separation time the liquid-liquid extraction is preferred. The radiochemical yield was 82% ± 6%. The activity concentration in the 56Co stock solution was ~4.5 MBq / ml.

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Separation of radioiodine by dry distillation process from irradiated elemental Te target

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Abstract – The yield of release of radioiodine from pressed elemental tellurium target and both sublimation and evaporation rate of target material were investigated under different experimental conditions.

Keywords – elemental tellurium target, dry distillation

I. Introduction

A separation of radioiodine by a dry distillation from irradiated TeO_2 target is widely used[1-6]. Until now, one work on studies on the separation of radioiodine by dry distillation from irradiated elemental tellurium target was published by Acerbi et. al.[7] in the seventies. In recent years the separation of radioiodine by anion-exchange and solvent extraction from metallic tellurium target was also investigated[8]. In the present work the dry distillation method of radioiodine from irradiated elemental tellurium target was studied.

II. Experimental

The thickness of pressed tellurium powder with a natural isotopic composition was about 320 mg/cm^2. Some of the targets were initially heated at a temperature 713 K and pressure 0.7 Pa by 10 minutes before an irradiation. The irradiation condition was 20 nA as proton of energy 60 MeV for 2-3 h at the Cracow AIC-144 cyclotron. Target after irradiation was placed in a quartz tube of 10 mm diameter. Performed two series of experiments: under reduced pressure – 0.7 Pa and at a flow of Ar - 15 cm^3/min. The duration of each experiment was 40 minutes after reaching a given temperature.

III. Results and discussion

The yield of release of radioiodine from target matrix and evaporation rate of target material were investigated under reduced pressure – 0.7 Pa for temperatures from 713 K to 823 K. The yield of release of radioiodine was about 65% for 713 K and reached the value 95% for 823 K. The tellurium loss in the separation conditions was 4% for temperature 713 K and increasing to 44% for 823 K. In the second series of experiments initially heated targets and Ar gas flux of 15 cm^3/min were used for temperatures lower then the melting point – 723 K of tellurium from 623 K to 713 K. A low yield of separation radioiodine was observed in the temperature range 623 K to 673 K. With the increase of process temperature to 713 K a significant increase of yield of dry distillation of iodine took place and reached the value about 65%. The tellurium losses in this temperature were from 3 to 4%. Comparing the obtained results of yield of release of radioiodine for targets with and without initially heating showed that the yield was higher by 12% at 698 K and about 17% at 713 K for targets without heating. There was no significant differences between the target material losses for a given temperatures. The sublimated tellurium condensed from 656 K to 575 K but the adsorption zone of radioiodine starting from 575 K to 320 K at the flow conditions. Due to overlapping the peaks the loss of radioiodine was 1,5%.

IV. Conclusion

The studies allowed to obtain the following conditions for the separation process: yield of release of radioiodine 85% (target without initially heating) and 65% (target with initially heating) at temperature 713 K. It follows that to achieve 85% efficiency of release will be required to reestablish the original properties of the tellurium powder. The sublimation of tellurium was 3-4% from initial mass. In comparison with TeO_2 target the yield of separation of radioiodine is lower then 14% (for the 40 minutes experiment). Taking into account the adsorption temperature of radioiodine it is possible to transport radioiodine to attached trapping device by heating the part of quartz tube beetwen end of furnace and attached trapping device at temperature above 385

K. Based on the results further tests are planned.

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An Automated Production of 64Cu on 18/9 MeV cyclotron

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Objectives: The reaction route 64Ni(p,n)64Cu is very popular for the preparation of 64Cu because its entrance channel is accessible at low energies and yield of reaction is quite high. The objective is to develop a module for automated production of 64Cu at BIONT site.
Methods: Electrodeposition was performed in galvanostatic mode at current 10–100 mA. Electroplating was accomplished within 1.5–12 h. A thickness of Ni layer was determined
by calculation after weighing of Ni on a disk for 1.13 cm2 area. A quality of surface layer was examined by SEM (scanning electron microscope). COSTIS target station was installed at the end of theexternal beam line of the IBA Cyclone 18/9 cyclotron. The target station has been equipped with 300 μm Nb window foil in the front of the target to degrade energy to energy
for nuclear energy less than 14 MeV. The homogeneous beam with area of 1.2 cm2 with of 5 μA proton and energy less than 14 MeV was applied. An automated separation module for isolation of 64Cu equipped with PLC SIMATIC S7-1200 controller has been developed. The quality of 64Cu was checked by gamma spectrometry and chemical purity (64Ni) was determined by ICP-MS.
Results: The target was prepared by electrodeposition of nickel proceeds according to the Davison and Harrison [1] and Philip and Nicol [2] in ammoniacal solution at pH 9. Target was irradiated with current of 5 μA in the COSTIS target station. 64Cu production rate for 100 mg 64Ni of 99.09 % purity (ISOFLEX) on gold target was 104 MBq/μAh. The separation of 64Cu from the target was achieved by anion exchange chromatography with HCl as elution solution. The chemical purity of 64Cu product was checked by ICP MS and concentration of 64Ni was less than 2 ppm.
Conclusions: The procedure developed has been successfully applied for the routine production of no-carrier-added 64Cu from enriched 64Ni generated by the reaction 64Ni(p, n)64Cu using a cyclotron 18/9. An automated separation module with PLC SIMATIC S7-1200 remote control has been constructed for the preparation of 64Cu with good radionuclide and chemical purity.
Research Support: This work was supported by the Slovak Research and Development Agency under the contract No. VMSPP-0075-09.

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Radiochemical and cross section studies for the production of the therapeutic radionuclide ^{193m}Pt

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The radionuclide ^{193m}Pt ($T_{1/2}$ = 4.33 d) decays by highly converted isomeric transition emitting about 33 Auger electrons. It is of great potential interest in internal radiotherapy because of its suitable decay properties. So far the specific activity of ^{193m}Pt produced has been rather low due to the use of the ^{192}Pt(n,γ)-process at a reactor. The aim of this work was to measure the excitation function of the ^{192}Os(α,3n) reaction. Furthermore, the yield and the specific activity of ^{193m}Pt produced was determined.
The dissolution of osmium, its conversion to OsO_4 vapor and trapping in KOH solution for electrolytic preparation of thin target samples of highly enriched ^{192}Os on Ni as well as the complete radiochemical separation of radioplatinum including ^{192}Os recovery were optimized.
The excitation function of the ^{192}Os(α,3n) ^{193m}Pt reaction was measured up to 39 MeV using the stacked-foil activation technique with 99.65 % enriched ^{192}Os targets at the Brussels Cyclotron. The ^{nat}Ti(α,x) ^{51}Cr reaction induced in Ti foils was used to monitor the incident beam intensity of about 100 nA. After a clean separation of radioplatinum, the radioactivity of ^{193m}Pt was measured via X-ray spectrometry using the Pt $K_{\alpha 1}$ X-ray line of 66.8 keV. The cross section of the ^{192}Os(α,3n) ^{193m}Pt reaction reaches a value of about 1.5 b at the maximum at about 35 MeV. It was also calculated theoretically using the compound-precompound nuclear model codes TALYS and STAPRE. The calculated values are consistent with the measured data. The optimum production of the ^{193m}Pt radionuclide via this route was found in the range 30-38 MeV; the integral yield of ^{193m}Pt amounted to 10 MBq/μAh. Its radionuclidic purity was found to be > 99% and the determined specific activity amounted to 1 GBq/μg Pt, which is by about 10^3 higher than in reactor production. Thus basic information on the production of high-quality ^{193m}Pt for therapeutic application is provided.

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Nuclear spallation reactions in chromium, yttrium and terbium with 386 MeV neutrons

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Neutron-induced reaction cross sections serve as a comprehensive nuclear database for estimating residual radioactivities in accelerator facilities. These data are also important in the field of cosmochemistry for deciphering the cosmic-ray irradiation history. However, neutron cross sections in the energy range above 100 MeV have scarcely been measured experimentally except for those from few targets; C, Cu, Pb, Bi and so on. Those obtained from calculation codes are utilized and proton cross section data are also utilized on the basis of the assumption that neutron cross sections in higher energies than 100 MeV approximately equal to proton ones in the same energy range.

In this work, we measured reaction cross sections of radionuclides produced through nuclear spallation reaction from Cr, Y and Tb induced by neutrons at 386 MeV, which have never been reported. The irradiations were carried out using neutrons produced through Li-7 (p, n) reaction at N0 beam line in the Research Center for Nuclear Physics (RCNP), Osaka University. To estimate quasi-monoenergetic neutron induced cross sections, the target stacks of Cr, Y and Tb were irradiated on the two angles of 0 and 25 degrees for the axis of the primary proton beam. The yields of the spallation products were measured by gamma-ray spectrometry. Neutron cross sections were estimated by subtracting the yields produced in the samples placed on 25 degree from those of 0 degree to correct the contribution of the low energy tail in the neutron spectrum. The results obtained in this work will be compared to the cross section data for the same target materials with 287 MeV neutrons in our previous work. Since proton-induced cross sections for the same targets in the same energy range are also measured, neutron cross sections obtained can be compared to the proton ones.

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Nuclear and radiochemical study of production and utilization of radioactive astatine isotopes in the 7Li+natPb reaction

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An alpha radioactive nuclide 211At with a half-life of 7.2 h is a prospective candidate for utilization in targeted alpha radiotherapy. In a general way, 211At is produced through bombardment of a bismuth target with 28 MeV helium ions in the 209Bi(alpha,2n)211At reaction because of the high yield required for therapeutic purpose [1]. However, the nuclear reactions using lithium ion beams, 6,7Li+Pb and 6,7Li+209Bi, provide the possible production routes of 211At. Excitation functions have been extensively measured for the 6,7Li+ 209Bi reactions to study the reaction mechanism involving complete fusion and breakup reaction of weakly bounded nuclei 6,7Li [2-4]. For 7Li+natPb, however, only reports on production of astatine isotopes 207-210At have been available for radiotherapy [5]. Therefore, we have measured excitation functions of 208-211At in the reaction of 29-57 MeV 7Li+natPb at the tandem accelerator of JAEA-Tokai. The cross sections of radioactive products were determined by alpha- and gamma-ray spectrometry. The cross sections of 211At below 45 MeV are large compared with those of the other astatine isotopes 208-210At. The experimental excitation functions of astatine isotopes have been compared with a statistical calculation to study the reaction mechanism. Besides, a chemical separation of carrier-free radioactive astatine isotopes from an irradiated target has been studied with a dry-chemistry method. Details will be shown in the presentation.

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Polonium Evaporation Studies from Liquid Lead-based Alloys

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Liquid lead and liquid lead-based alloys are currently in discussion for application as spallation target material and coolant in neutron sources and Accelerator Driven Systems (ADS). The main disadvantage of lead-based alloys is the formation of Po-210 by nuclear reactions. This radionuclide is an alpha-emitter with a half-life of 138.8 days and therefore of high radiological concern due to inhalation or ingestion in case of its release to the environment. The production of this polonium isotope is especially pronounced in bismuth containing alloys such as lead bismuth eutectic (LBE), which is foreseen as spallation target material and reactor coolant in the ADS prototype MYRRHA. To reduce the hazard resulting from polonium release, pure lead and lead gold eutectic (LGE) are considered as alternative liquid spallation target materials and reactor coolants. Though the production of polonium in these materials is much lower compared to LBE, still considerable amounts are formed. Therefore it is crucial for the licensing of such facilities to study the release behavior of polonium from these materials. In the present study we have investigated the evaporation behavior of Polonium from pure lead and lead-gold eutectic using the transpiration technique. The results are compared with earlier measurements of polonium evaporation from LBE.

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Elemental analysis of rivers, marshes and ground water in Thi Qar region, Iraq

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The elemental analysis of the samples collected from marshes water, river water and ground water in Thi Qar region of Iraq are done using the Energy-dispersive X-ray fluorescence technique (EDXRF). The water samples are collected from Al-Hammar marshes, Central marshes, Tigris river and Euphrates river. The residues obtained after drying ground water are analyzed using the EDXRF spectrometer is consisting of 42Mo-anode X-ray tube equipped with selective absorbers as an excitation source and a Si (Li) detector. The elements observed in various samples are 20Ca, 26Fe, 29Cu, 30Zn, 33As, 35Br, 38Sr, 82Pb and 92U. The pH value of water samples from various locations of marshes, rivers and ground waters are in general exceeding ~7.9 value, which indicate the general alkaline nature of waters. The average value of leftover residue obtained after drying the water samples from Al-Hammar marshes is ~ 4.8 g/L, which is about twice of that of Central marshes. The average value of total residue obtained after drying the water samples from Euphrates river is ~ 3 g/L, which is about four times of that obtained from Tigris river water. The Electrical Conductivity (EC) values measurements for these samples also infer the similar conclusions. The 20Ca concentration in Tigris and Euphrates river waters is similar to that observed in Central marshes and is about half of the value in Al-Hammar marshes. The 20Ca concentration in Tigris river water constitutes ~ 6 % of the leftover residue and that in Euphrates river water constitutes ~2.3 % of the total residue, a value similar to that observed in water from the marshes. The 38Sr concentrations in the Tigris and Euphrates river waters are in the range 1390–1863¬ μg/L and 2519–5691 ¬¬μg/L, respectively. The 38Sr concentration in Euphrates river is similar to that observed in the marshes water and that in Tigris river is lower by a factor of 8. In Al-Hammar marshes, 82Pb is observed with concentration ~ 60μg/L and the concentration of 34As is estimated to be in general below 5 μg/L. In Central marshes, 28Ni and 29Cu are also observed with average concentration of 40 and 28μg/L, respectively. Average value of concentration of the trace element 92U in both the marshes is below 10μg/L. The 35Br concentration in various samples collected from Al-Hammar marshes (average value = 1108 μg/L) are in general higher compared to those from Central marshes (average value = 328 μg/L). The 35Br concentration in various samples collected from Euphrates river (average value = 676 μg/L) are considerably higher compared to those from Tigris river (average value = 47 μg/L). The presence of significantly high 35Br concentration in the river water is a matter of concern in case it is used for drinking purpose after treatment. The normally present bromide anions can generate bromate anions in the water purification based on ozonation or chlorination or ultra violet radiation. In case the Br concentration of water is very high, it is likely that bromate ions will also be formed to significant extent. The water from Tigris river is safe for drinking water supply schemes after treatment and Euphrates river is not safe this purpose due to high bromine content.

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Investigation of uranium contamination in ground water of southwest Punjab using EDXRF technique

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The Energy-dispersive X-ray fluorescence technique (EDXRF) is used for elemental analysis of the samples collected from the ground water and the canal water in the Bathinda district of Punjab state, India. The residues obtained after drying the water samples are analyzed using the EDXRF spectrometer consisting of 42Mo-anode X-ray tube equipped with selective absorbers as an excitation source and an Si(Li) detector. The maximum concentrations of 35Br, 38Sr and 92U elements are observed to be 5543, 6165 and 212 μg/L, respectively, in shallow ground water samples and 20, 200 and 5 μg/L, respectively, in the canal water samples. The observed concentrations of these elements show positive correlation with the Total Dissolved Salt (TDS) content of the ground water. To investigate the flyash from the coal-fired thermal power plants as a possible source of ground water contamination, the water samples collected from the surroundings of the power plants and the flyash samples are also analyzed. The elemental analysis rule out the possibility of flyash as a possible source of ground water contamination. The canals reaching the Malwa region are based on Sutlej river and the canal water is used extensively for irrigation. Samples collected from different locations of industrial waste water drains sinking into Sutlej river near Phagwara and Ludhiana were also analyzed. It is concluded that the industrial drains lead to uranium contamination in Bathinda. The presence of uranium in the ground water in the southwest Punjab due to weathering of granite rocks with high radioactivity content (uranium concentration ~8 ppm) at Tosham hills (distance ~ 200 km from Bathinda) is rather unlikely in the absence of very supportive conditions, viz., large water body with abnormal pH values and ground water flow from Tosham hill region to southwest Punjab. As the uranium contamination is occurring over vast regions of ground water, the source is likely to be not localized one. Southwest Punjab consists of Sutlej-Ghaggar plain. The soils in the region have largely developed on alluvium deposits up to depth of thousands of feet and evidences like high concentration of soil radioactivity or radon also do not favour existence of natural uranium ore deposits in Bathinda. It is a rich agricultural area cultivated with the help of extensive irrigation made possible by the canal water from the Sutlej river. The soils in the region are calcareous and contain uranium ~ 3-5 ppm. The ground water level is in the region is in general shallow. The irrigation schemes are mainly based on extensive network of canal water. The irrigation water percolating through the soil dissolves carbon dioxide produced from the plant root respiration and the microbial oxidation of the agricultural matter and results in carbonic acid. The carbonic acid reacts with the calcium carbonate (calcareous soil) to produce bicarbonate, which leaches uranium from soils and sediments to the ground water. Further, the use of agricultural additives like phosphates fertilizers (uranium concentration ~ few tens of ppm) enhances the uranium and chemical concentrations in the ground water. Due to minimal use of ground water, the chemical contamination in ground water is increasing.

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Sorption of Cadmium, Nickel, Caesium and Strontium to a Laterite Soil: Application of Linear Additive Model and Surface Complexation Modelling

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Laterite soil obtained from Lagos, Nigeria was comprehensively characterised to determine the organic matter content, cation exchange capacity, surface area, and of mineralogical composition. The laterite was constituted mostly of Quartz (47.2 %), Kaolinite (42.6 %), Goethite (7.2 %), and hematite (2.1 %).

Batch sorption experiments for Cd, Cs, Ni & Sr to the laterite were performed from pH 5 to 9, and to a synthetic laterite soil prepared using commercially available analytical grade minerals. The individual sorption profiles of the metals with the pure minerals were obtained, as well as those of mixtures of the minerals in incremental complexity, a Linear Additive Model (LAM) has been used to calculate the theoretical Rds of the mixtures from the individual Rds. Potentiometric titrations of the solids were carried out to obtain log K values and proton exchange capacities, and these have been applied to surface complexation models (SCM) using the speciation software JCHESS, to predict sorption profiles.

The sorption profiles of the different metals with the natural and synthetic laterites have been compared to the profiles calculated with LAM and predicted with SCM. The sorption profiles obtained from the incremental complex mixtures allow for better understanding of differences between the experimental, calculated or predicted sorption profiles, if any.

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Interaction of Cm(III) with human serum transferrin studied by Time-Resolved Laser Fluorescence Spectroscopy (TRLFS)

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In case of an accidental release of radionuclides to the environment actinides can cause a serious health risk upon incorporation. There is only deficient knowledge about the chemical behavior and toxicity of actinides in man. With regard to the development of potential decontamination therapies, a detailed understanding of the mechanisms of relevant biochemical reactions is necessary.[1] Human serum transferrin is an iron carrier protein in the blood. It is folded into two lobes housing the metal binding sites for Fe(III).[2] In the normal blood serum, only 30 % of transferrin is saturated with iron, which indicates that there is a high capacity for the complexation of other metal ions.

Human serum transferrin is found to be contaminated by EDTA. In the present work a purification process was developed using size exclusion chromatography and centrifugal filters. After purification the complexation of Cm(III) with transferrin is studied at various pH values and transferrin concentrations by time-resolved laser fluorescence spectroscopy (TRLFS). The results show that two different species are formed. In the pH range from 6.3 to 7.7 the spectra are dominated by the Cm(III) transferrin species I, displaying an emission band at 600.0 nm. The fluorescence lifetime of 97 µs correlates with a coordination of six water molecules indicating a threefold coordination mode with the protein and/or a synergistic anion such as hydroxid or carbonate at the binding site.[3] Above pH 7.7 the Cm(III) transferrin species II with an emission band at 620.3 nm is formed. The extraordinary bathochromic shift of 26.6 nm relative to the emission band of the Cm(III) aquo ion and the fluorescence lifetime of 221 µs confirm incorporation of Cm(III) at the transferrin binding site resulting in a 4-fold coordination via amino acid groups (Asp-63, Tyr-95, Tyr-188 and His-249) of the protein. The remaining coordination sites of Cm(III) are occupied by synergistic anions and water molecules.

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Application of Non-linear Heterogeneity-based Isotherm Models for Charactering Sorption of Cs and Se on Mudrocks

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It plays a very important role for characterizing sorption behaviors of cesium (Cs) and selenium (Se) on Taiwanese mudrocks to retard the migration of radionuclides from a radioactive waste repository. In this study, two non-linear heterogeneity-based isotherms, Langmuir-Freundlich model (LF) and generalized-Freundlich model (GF), were applied for the evaluation of the sorption characteristics of Cs and Se on Taiwanese mudrocks. The batch sorption experiments were carried out and the experimental data were simulated by LF and GF heterogeneity-based isotherm models. In addition, the results showed that both of the LF and GF models could fit the experimental data more perfectly than the Langmuir one. The heterogeneity of sorption behaviors for Cs and Se could be well characterized by the LF and GF models from the root mean square error (RMSE) calculation and plot of the affinity spectrum. The results demonstrated that the sorption mechanism of Cs and Se on mudrock is quite different and Cs sorption is more heterogeneous and complicated than that in Se.

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Experiments to demonstrate chemical containment: Solubility under the cementitious conditions of a repository in the UK

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One option for the long-term management of intermediate level radioactive wastes is deep geological disposal. The waste is grouted with cement in steel canisters for disposal; after closure, the repository may be backfilled with more cement. The cement porewater is expected to be of high pH due to cement dissolution and low Eh due to canister corrosion. This work describes a series of long term experiments to demonstrate precipitation and solubility limitation under these conditions.

The experiments are expected to last 4 years and interim results are presented here. The elements included are Cs, Ni, Eu, U and Th, I as iodide and Se, as SeO_3^{2-}. Four solutions are being tested, NaOH, saturated $Ca(OH)_2$, NRVB (a limestone-rich backfill) -equilibrated water and cellulose degradation products (CDP) in NRVB. Additional experiments in CDP are being carried out in the presence of reducing agents (metallic Fe and $Na_2S_2O_4$). Solubility is assessed from both under- and oversaturation directions.

After one year of periodic sampling, steady state has been reached for U and Ni. $Ca(OH)_2$ is a good surrogate for the behaviour of radionuclides in NRVB, showing similar results to the reference backfill. CDP increase the solubility of Ni and U by several orders of magnitude, whereas Th and Eu are less affected. The solubility of Se is clearly conditioned by the availability of Ca in solution, and there is no evidence of a decrease in the concentration of Cs or I by co-precipitation. XRD analyses of precipitated solids show poorly crystalline phases in the oversaturation experiments. Only incipient crystallinity was observed for undersaturation, which could explain the differences observed in measured solubility between the two sets of experiments.

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Attachment of APTES ((3-aminopropyl)triethoxysilane) to silica for sorption and selective removal of radionickel from solution

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The main aim of this research is to design and develop a novel class of selective sorbents or sequestrating agents for various actinides, fission products, heavy metals and groundwater contaminants using a bespoke modular design of solid-supported polymers and containment-specific ligand groups.
Our current research has seen us attach the ligand known as APTES straight to the surface of silica gel (mean particle size; ca. 70 μm) and high surface area fumed silica (mean particle size; ca. 0.007 μm).
By utilising two different silica types, the authors have been able to investigate the efficiency of ligand attachment to different types of silica. Following successful sequestration of inactive transition metals including cobalt, nickel, copper and zinc, we have extended our research by using the radioactive isotope of nickel-63.
Using a range of concentrations from 2.5 ppm to 80 ppm, the silica attached APTES has successfully sequestrated Ni-63. To make the investigation more realistic and be able to relate it to a real case scenario, competitive ions (in this case sodium and calcium) were added to further the study. It has been shown that the nickel sequestration is not as affected by the addition of these ions as one might expect. Sequestration is still observed at a similar level to deionised water.
Rd's for the sequestration of Ni-63 from deionised water range from 4 x 104 ml/g to 1.2 x 107 ml/g compared to 5.3 x 104 ml/g to 7.9 x 105 ml/g for competitive calcium in solution and 1.2 x 105 ml/g to 7.3 x 106 ml/g for competitive sodium sequestration. Isotherms have also been produced across a pH range from 5.01 to 6.80 before addition of the material, to a final pH of 6.90 to 9.49 depending on the original concentration and competitive ions in solution.

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Chemical Decontamination, at Field, after 137Cs Accident at Goiania, Brazil

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In1987 to 1988, we were in charge of chemical decontamination of different materials, from soils, roads, hospitals, schools, houses indoor, house-wares and roofs to people, and their urines and feces, and water, highly contaminated with 137-Cs from a radiotherapy device (1300 Ci or 48,1 TBq), robbed from a deactivated hospital in demolition, in Goiania. The source structure we decontaminate weighed 304 Kg, the complete device weighed about 800 Kg, and could not have been put in a hand car, by 3 people, as was published. People dismounted the device in a junkyard, contaminated a lot of areas and materials in three junkyards, and took most of the 137-CsCl source to their homes, fascinated with the phos-phorescent blue light. When people were sick and burnt, they washed out the materials of original source , to the garden. After 25 years it is important to present and discuss the accident and the chemical solutions to the problem of sprayed CsCl from the original source, by common people, water and dust, due to high water solubility and chemical interactions with clay minerals and K-containing materials. Some information about the chemical decontamination work was published in the CNEN-IAEA Goiania Report, based in our internal report (1988). In NRC 7 we intended to present our work, but it was not possible to participate of the conference. In the present work we present and discuss chemical decontamination of different materials at field. Very often it is necessary to attack chemically the materials to let Cs+ available. The best ion exchange material for Cs+ is FeKFe(CN)6 (K Prussian Blue, or KPB) and not K4Fe(CN)6 (PB) used in “Radiogardase” capsules. Al3+ ions coagulate coloidal materials, H+ attack oxides and carbonates, K+ substitute Cs+ and KPB exchanges K+ by Cs+. To liberate Cs+ from dust, soils, cements, the best solutions are KAl(SO4)2 solutions, acidified with HCl. For granites it is better to use HF solutions, to remove Cs from fluorspat. Urine and feces were decontaminated with Resin/KPB. Resin/KPB is effective to decontaminate Cs+ hands and feet, instead of ingesting Radiogardase capsules, waiting Cs+ to go in the blood stream to be exchanged at the intestines. We produce KPB at field, in suspensions or supported in cation exchange resins, in wool towels, is EPU foams and in t-shirts. Published reports with decontamination methods to decontaminate materials and a city (Harshaw Report) , with NH4Cl or NH4NO3 were too light and slow. Process had to be much faster and effective, and had to take in account the chemical composition of every material and provide ion exchange to remove Cs+ ions.

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Assessment of present and future radioactive contamination at global scale

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As a result of plus than two thousand nuclear tests performed since 1945, two war actions and few accidents in nuclear reactor, it does already exists a radioactive contamination at global scale. This contamination has been accumulated mainly in marine sediments, because sea is about 80% of planet surface, and solid fission products released by nuclear explosions are transported by wind first and then by rain to liquid and solid portions on earth, while gaseous fission products are diffused in the atmosphere. In this way, heavy, high yielding fission products, such as Cs-137 get marine sediments, where they are found mixed with heavy radioactive natural elements such as U, Th and their also radioactive decay products. But since alkaline are so abundant on earth, it is also found natural radioactive isotope K-40, 0.0118% of isotopes forming K element, with half life 1.28x10E9, in the time range of planet age and elements origin. So, one easy way to assess both the importanceand evaluation of radioactive contamination at global scale, should be to establish a radioactive contamination factor (RCF) as a percentage of Cs-137 contaminant radioactivity in marine sediments, compared to K-40 natural radioactivity present from the very beguinning of earth, both expressed as Bq per gram of sediment. In the frame of these rather general considerations a research project has started in Mexico whose varied purposes are: to characterize sea regions in both vast litorals in the country, by its natural K-40 radioactivity present in sea salts, as well as to determine if Cs-137 traces produced by recent radioactive contamination have reached their coasts. Also, to assess the Cs-137 traces already present in marine sediments as a result of more than two thousand nuclear test explosions performed till now, rather than contamination produced by four main accidents in nuclear reactors during the last 60 years. It is proposed a radioactive contamination factor obtained from samples taken up in both coasts during the last two years, which it should be very useful in two ways: first to assess the real danger of radioactive contamination already present by comparing with natural radioactivity, and second to assess the growth, equilibrium or drop of radioactive contamination , given that nuclear energy is still the best option to solve the large energy demand foreseen in the near future.

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Trivalent Actinide/Lanthanide Sorption under Saline Conditions

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Sorption processes on mineral surfaces play an important role in the retardation behaviour of radionuclides and have to be considered in performance assessment calculations. However, almost all available experimental data are restricted to low ionic strength conditions. Saline conditions are not restricted to solutions relevant to rocksalt formations. Elevated salt concentrations up to 6.5 M are also found in sedimentary rocks which are under investigation within nuclear waste disposal programs.

In the present work, experiments have been performed with quartz and illite. In all cases sorption of Eu(III) radiolabelled with Eu-152 was studied in batch experiments, where ionic strength was varied up to 5 M NaCl. Experiments with Cm(III) and the time resolved laser fluorescence spectroscopy (TRLFS) are directed to obtain detailed information on the metal ion speciation at the mineral surfaces as a function of ionic strength.

In all systems we observed a decreasing sorption with increasing ionic strength. While for quartz only inner-sphere surface complexation is expected for Eu(III), outer sphere sorption at low pH must be considered for illite.

Eu(III) sorption to illite was calculated by a slightly modified well established non-electrostatic sorption model, developed and tested so far only for lower ionic strength conditions (2SPNE/CE). The Pitzer formalism was used to calculate activity coefficients for aqueous species. Model calculations for different ionic strengths yield fair agreement with the experimental data. This finding suggests that the shift in sorption edges with increasing ionic strength is basically due to a variation of ion activities and a concomitant change in aqueous metal ion speciation.

TRLFS experiments with Cm(III) indicate no significant differences for sorbed inner-sphere Cm(III) complexes at low and high ionic strength conditions. Our preliminary study indicates that the modified non-electrostatic 2SPNE/CE model can be applied to predict sorption of trivalent actinide ions also for elevated ionic strength conditions at least in NaCl dominated solutions.

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Structural studies of actinide-peptide complexes

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In the wake of the Fukushima accident assessing the mechanisms governing radionucleide impact on the environment (particularly the biosphere) is of great importance.
Most data available on the interaction of actinides with biological systems are based on physiological or biokinetic measurements, with scarce information on the microscopic factors such as structure of the actinide coordination site within biological molecules (proteins, peptides...). These biomolecules are essentially responsible for specific migration pathways towards various organs in vivo. This structural data is essential for developing the understanding structure, function and affinity interdependence. Here-in we describe an approach for structural elucidation of several uranyle and europium (used as a trivalent actinide surrogate) biological complexes combining both experimental and theoretical techniques.
the discribed approach overcomes the intricacy of actinide chemistry combined with the complexicity of protein tertiary structure by using biomimetic peptides. The chosen peptides cover a wide spectrum of structural as well as functional (amide, carboxylate et phosphonate…) properties. Isothermal calorimetric titration provided primordial thermodynamic information (affinity constant, stoechiometry), additional functional and structural analysis techniques (IR,TRLFS, EXAFS) led to elucidate the complexe's structure. In parallel DFT calculations have been used to optimize the proposed structures and fit the experimental data.

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Iodine - 129 in water samples from Germany

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The natural abundance of 129I has been changed through anthropogenic activities, such as nuclear weapon testing, nuclear accidents and reprocessing of nuclear fuel. Mainly as a con-sequence of continuous releases of 129I from the nuclear reprocessing facilities in La Hague (France) and Sellafield (UK), 129I is in a state of disequilibrium in all environmental compart-ments in Western Europe.
It is emitted in gaseous and liquid form, with most of it as liquid waste which flows into the North Sea [1]. From there it is reemitted to and distributed in the atmosphere, and precipitates as wet or dry deposition [2].
In this project we analyse precipitation samples from 10 sampling locations and river water samples from 15 sampling locations all over Germany. The concentrations of 129I and 127I are determined, as well as their isotopic ratio.
Iodine is extracted from the water by ion exchange resin. 127I is analysed using inductively-coupled plasma mass spectrometry (ICP-MS) and the isotopic ratio is determined via accel-erated mass spectrometry (AMS).
Results of the 127I analyses reveal slightly higher concentrations for the river water samples (1.7 to 15.9 ng/g) than for the rain water samples (0.6 to 9.8 ng/g). Concentrations of 129I range between 17 to 2000 fg/kg (rain water) and between 17 to 350 fg/kg (river water), respectively. For the precipitation samples (14·10-9 to 650·10-9), as well as for the river water samples (3·10-9 to 40·10-9), the 129I/127I ratios are at least 3 orders of magnitude higher than the natural equilibrium isotopic ratio of 1.5·10-12 [2].

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Archaeometry with INAA at the Research Reactor TRIGA Mainz

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At the research reactor TRIGA Mark II of the Institute for Nuclear Chemistry of the Johannes Gutenberg-Universität Mainz, Germany, the interdisciplinary field of archaeometry is studied by instrumental neutron activation analysis (INAA). Currently the work is concentrated on three different projects in archaeological context: Provenance analyses of ancient Roman limestone findings in the Rhine-Moselle region, determination of the composition of glass beads of the late La Tène era, which were excavated in southern Bavaria, and the analysis of sinter profile from ancient Mediterranean aqueducts to interpret its layers as climate archives. For all samplings, a method with a low risk of contamination is applied. To get a wide spectrum of elements (chemical fingerprint), the irradiations and the gamma-ray measurements are adjusted to the sample composition.

For the provenance project ancient limestone quarries as well as interesting archaeological objects are prospected and systematically sampled. Chemical fingerprints measured by INAA combined with multivariate statistics are used to determine the origin of the objects to shed light on the complexity of logistics, transport and trade routes in former times.

Glass beads from an ancient ritual place near Oberammergau in Bavaria, Germany are analyzed via INAA to determine the glass type, coloring elements and trace elements. The lanthanide concentrations deliver information about the raw glass production and give archaeologists clues as to the origin of the raw materials. Measurements from X-ray photoelectron spectroscopy together with the results of the INAA analyses provide insight into the historic manufacturing process of the beads.

To garner a better understanding of extreme weather event risks and the frequency of earthquakes in the present, the knowledge of the occurrence of these events for long periods of time is necessary. In the initiated project, sinter layers from ancient aqueducts are used as climate archives with proper chronological resolution.

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Characterisation of Radioactive Scales (NORM) Produced by the Onshore Oil and Gas Industry in the UK

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The occurrence of radioactive scales in oil & gas production facilities is a major concern for the petroleum industry. Activities exceeding 1KBq/g radium-226 and 30KBq/g lead-210 have been reported in solid scales from the UK sector. When progeny are taken into account, total activities levels may exceed the UK threshold for Low Level Radioactive Waste (LLW) disposal. Increased radiation risks to operators and general public are now being addressed by revised legislation; however the additional costs of waste management are yet to be fully assessed. Much of the work carried out to date has focussed on offshore NORM where the majority of arisings are disposed to sea. However, onshore treatment, where sea disposal is not an option, poses a serious challenge.

The aim of this project is to establish the nature, extent and distribution of NORM in UK onshore oil and gas fields and to devise an effective NORM inhibition methodology for minimising future health, environmental and economic impacts. Radiological surveys of production equipments are being undertaken and radiometric and isotopic analysis are used to characterise radioactive scale deposits in representative UK onshore oil & gas fields. Methods for inhibiting radium isotope co-precipitation with common mineral phases such as barite and celestite, are being investigated by nucleating crystals in a counter diffusion system. Preliminary results are described.

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Structural Transformations in Metallic Iron under the Action of External Irradiation

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Using several modes of Mossbauer spectroscopy, after effects have been studied of irradiating metallic iron with 1) thermal neutrons (fluence of 1.8·E24 n·m-2); 2) protons (energy, 6.0/2.0 MeV; fluence, 1.0·E22 p·m-2); 3) deuterons (energy, 9.0/7.3 MeV; fluence, 1.0·E21 to 1.0·E22 d·m-2); 4) α-particles from a 238Pu source (energy, 5.5. MeV; fluence, 2.5·E19 α·m-2); 5) 12C- and 14N-ions (energy, 47.2/0 and 58.8/0 MeV, respectively; fluence, (1.6 to 8.2)·E19 ions·m-2); 6) 6.1 to 8.8 MeV α-particles and 208,212Pb, 212,216Po recoil nuclei from a 228Th-source (energy, 0.11 to 0.17 MeV), the total fluence being 4.5·E18 particles·m-2.

The experimental data obtained in the study enabled various types of external radiation to be correlated as to their radiation damage, the effect on the structure-, phase composition- and corrosion resistance properties of metallic iron.

After irradiating with neutrons, protons and weak deuterons (beam currents of less than 5 μA), it is only the magnetic superfine structure, which is characteristic of alpha-Fe, that has been observed in the experimental spectra.

Irradiation with intensive beams of deuterons (beam currents, 10 to 15 μA), α-particles , 12C- and 14N-ions leads to a structural disordering of the alpha-Fe lattice and to the emergence of the gamma-phase on the surface of foils and in the near-surface area: a single component, which is 2 to 3 times wider as compared to the magnetic sextet lines: is a result of local heating of the lattice to high temperatures with subsequent recrystallization from the “molted" volume.

Irradiation of iron foils with recoil nuclei (combined with α-particles) provokes corrosion processes and is accompanied by an intensive oxidation of the metal.

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THEREDA – a Thermodynamic Reference Database project

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Radioactive waste poses a serious danger to the environment. Consequently, many significant efforts are made to ensure the safe disposal of hazardous waste. In order to assess the long term safety of a repository, geochemical model calculations are used to analyze the performance of a system and predict the amount of radionuclides potentially mobilized from a repository. For reliable geochemical model calculations, a consistent, comprehensive and well maintained thermodynamic reference database is needed. The main objective of THEREDA is to establish a comprehensive and internally consistent thermodynamic database for the geochemical modelling of near-field and far-field processes occurring in the different host rock formations under discussion. The THEREDA database is run by the following institutions:
GRS: Gesellschaft für Anlagen- und Reaktorsicherheit mbH, Germany
KIT-INE: Karlsruhe Institute of Technology, Institute for Nuclear Waste Disposal, Germany
HZDR-IRC: Helmholtz-Zentrum Dresden-Rossendorf, Institute of Resource Ecology, Germany
TU-BAF: Technische Universität Bergakademie Freiberg, Institut für Anorganische Chemie, Germany
AF-Consult: AF-Consult Switzerland AG, Switzerland
THEREDA is focusing on data for actinides, fission- and activation products, chemotoxic elements and the elements defining the matrix system, i.e. the system of oceanic salts and cement phases. Specific labels are assigned to the data included in THEREDA allowing for traceability and transparency. THEREDA database is organized in a way to assure that compatibility problems (e.g. arising from exclusively SIT or Pitzer compatible data) are managed. The database is handled by a central databank, and made available to potential users via internet. Software tools are supplied to translate THEREDA into input files for commonly used geochemical modeling codes (EQ3/6, PHREEQC, Geochemist's Workbench, CHEMAPP, ...).
For more information please visit the project webpage at www.thereda.de where more detailed information on the THEREDA project and contact addresses are given.

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Characterization of the Natural Organic Matter (NOM) by ultrafiltration and fluorescence in a groundwater plume contaminated with 60Co and 137Cs

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Natural Organic Matter (NOM) is a collection of molecules originating from the decomposition of built biomass, and also from exudates from biological activities. The constituents of NOM are poorly characterized molecules of various molecular sizes and functional groups that could affect the fate of radionuclides and other contaminants. In this work, groundwaters have been sampled near a contaminated area in the Canadian boreal shield and size-fractionated by ultrafiltration (5000 Da cut-off) to determine the associations of selected radionuclides (60Co, 137Cs) with colloidal-sized fractions of NOM. Solid phase extraction (SPE) was also used in tandem with fluorescence analysis of the fractions, to elucidate the changes in the chemical nature of the NOM. Fluorescence is a powerful tool that can track the optical characteristics of the NOM constituents in an Excitation-Emission Matrix (EEM). The EEM, in turn, is numerically decomposed into individual and independent components, defined as humic-like, fulvic-like, and protein-like.

Our results have revealed consistent trends over the years (2004-2010): an uncontaminated station had a small colloidal NOM content (typically <7% was below the filter cut-off), whereas the colloidal NOM was higher in the contaminated sites (typically 12-41% of the total). Cesium-137 was dominant in the colloidal NOM fraction (>95% of the total), whereas 60Co was mostly in the filtered fraction (70-90%). When the samples were submitted to SPE and fluorescence, a systematic removal of the protein-like NOM was found, without affecting the humic- and fulvic-like components. Cobalt-60 and 137Cs were affected only to a small extent, suggesting that these were associated with the humic- and fulvic-like NOM, and not the protein-like NOM. This finding is intriguing and unique, as the protein-like NOM was found only in the contaminated sites. Applications of fluorescence, as a new tool to this sampling, will be discussed further.

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Iodine-129 and iodine-127 in aerosols from Northern Germany

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I-129 ($T_{1/2}$=15.7 Ma) is produced both naturally and as result of human nuclear activities. Nowadays, the amount of anthropogenic I-129 in the atmosphere and in surface waters has continued to increase due to releases from the spent nuclear fuel reprocesssing plants, particularly in Western Europe, from the nuclear reprocessing plants La Hague (France) and Sellafield (U.K.). In the atmosphere, iodine exists in differnt species as inorganic gaseous iodine (I_2, HI, HOI), organic gaseous iodine (CHI_3, CH_2I_2, $CH_3CH_2CH_2I$, etc.) and particle associated iodine; their concentrations vary with location, season and climate. The speciation of I-129 in the atmosphere can also provide useful information about sources and transfer pathways.
In this work, we present the concentration of I-127, I-129 and the ratio of I-129/I-127 in aerosol samples, collected weekly during the year 2011 in Braunschweig, Northern Germany. Aerosols were extracted from the filters using an alkaline solution and separated from the matrix elements with chloroform. The samples were analysed for I-129 by accelerator mass spectrometry (AMS), and for I-127 by inductively coupled plasma mass spectrometry (ICP-MS).
The concentrations of I-127 and I-129 in aerosol filters range between 0.4-3.7 ng/m³ and 0.06-0.7 fg/m³, respectively. The I-129/I-129 ratios range between 0.7 x 10E-7 and 7.4 x 10E-7. In addition, we compare the distribution of I-129 with that of Kr-85 and Be-7. We discuss the effect of speciation of I-129 and stable I-127 in the environment, and the disequilibrium between these two isotopes.

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A Solid-State NMR Study of the Complexation of 109Cd with Isosaccharinic Acid

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The binding of 109Cd to isosaccharinic acid using advanced solid state NMR techniques was examined. The complexes, as well as ISA itself, were analysed at 3 different pHs (7, 10 and 13). Various solid-state NMR techniques were used. CP-MAS provided C-13 spectra of the complexes and the related dipolar dephasing method helped to determine the assignment of CH and CH2 groups. Slow sample spinning provided sufficient spinning side bands to allow the extraction of the CSA values. Experimental compared with calculated techniques were also carried out using Gaussian, Herzfeld Berger and Simpson programmes to look at differences in the spectra but also the CSA of the nuclei in the complex. The spectra of ISA at the various pHs were obtained first. Spectra were then obtained for the metal-ligand complexes. These spectra have enabled more detailed theories to be drawn up on the nature of the binding of radionuclides to polyhydroxylated carboxylic acids at varying pH.

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Generation of inorganic colloids in the chemical disturbed zone in the proximity of a cementitious repository

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The current concept for the disposal of intermediate- and low-level waste in the UK involves the placement of grouted waste confined in stainless steel canisters in a cementitious repository deep underground. While the cement will buffer the porewater to a highly alkaline pH, contributing to the retardation of radionuclides by precipitation, this could potentially create an alkaline plume moving from the repository into the host rock. The alkaline cement leachate can react with the host rock, promoting dissolution of some mineral phases, precipitation of new phases and the generation of colloids that may affect radionuclide movement within the chemically disturbed zone of the host rock.

The aim of the present work is to study the generation of inorganic colloids from a generic host rock in contact with an alkaline plume. An intact core of sandstone has had an artificial "young" cement leachate (pH 13.10) under CO_2-free conditions pumped through it. The solution at the outlet was then sequentially ultrafiltered in-line through a rig fitted with

12 µm, 1 µm, 30 kDa and 1 kDa membranes. After passing more than 500 pore volumes of the young cement leachate, the imaging of the filters by Scanning Electron Microscopy (SEM) in combination with X-ray microanalysis showed no evidence of the formation of inorganic colloids under these experimental conditions. On the other hand, analysis of the outlet solution by ICP-MS indicates that there is significant dissolution of some mineral phases present in the sandstone, however no difference were found between the elemental composition of the solution before and after ultrafiltration. Additional tests with artificial colloids indicate that silica and iron oxides colloids are unstable in "young" cement leachate.

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Uranium in ground water samples from Anthemountas Basin, Northern Greece

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The activity concentrations of 238U and 234U have been determined in groundwater samples of deep wells in Anthemountas river basin, Northern Greece. The analysis was performed by alpha spectroscopy after pre-concentration and separation of uranium by cation exchange and finally its electro-deposition on stainless steel discs. Analysis by gamma spectroscopy was also performed. The uranium concentrations in the region of study and during winter period varied strongly between 9 and 42.6 ppb. These variations in concentrations of uranium are correlated with the different geological formations and water flow paths in the study area. In the case of Anthemountas river basin the available data revealed a very complex system of aquifers with significant extent in both lateral and vertical sense. The study area is covered with sediments deposits consisting of: i) Valley deposits: sandy clay, ii) Lower terrace system: gravels and sand under a clayey cover, iii) Upper terrace system: grits and pebbles with loam or sandy clay, iv) Fans of different age, v) Red clay series: red to brick red, silty clays with mica and calcareous concretionary bodies, while the area to the north and south are covered by vi) Limestones recrystallised, Granodiorite, viii) Gabbro and ix) Dunites and peridotites. The hydrogeology varies significantly for the area of interest due to the anisotropy of the sediments of the basin. Their behaviour is driven mainly by their primary porosity and the secondary porosity caused from the faulting tectonics making it easier for water flow in certain areas. From the radiological point of view, using ground waters for human consumption would result in significantly increased radiation doses and their utilization for irrigation purposes would imply an excessive radiation exposure to population, in cases of high uranium concentrations.

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Time lag between the tropopause height and ^{7}Be activity concentrations on surface air

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The objective of this study is to define the time-lag between the elevation of tropopause and the concentration of 7Be in near surface air. The concentration of 7Be at surface air has been determined in the region of Thessaloniki, Greece at 40°N, over 52 weekly measurements covering the year 2009, a year of a deep solar minimum and of maximum concentration of 7Be, where any fluctuation due to meteorological and seasonal variations are easily revealed.

Sampling of 7Be aerosols was carried out by Staplex high-volume air sampler with glass-fiber filters, and a regulated airflow rate of 1.7-1.92 m3 min-1 (60-68 ft3 min-1). The length of each collection period was 24 h. All samples were measured for 7Be activity using an HPGe detector (42% relative efficiency). The tropopause height time series of daily values was obtained from the NCEP/NCAR Reanalysis data.

The positive correlations between the monthly activity concentrations of 7Be and the tropopause height (0.94, $p<0.0001$) as well as between 7Be concentrations and the temperature T (°C) ($R=0.97$, $p<0.001$), confirm that the increased rate of vertical transport within the troposphere, has as a result the descent to surface of air masses enriched in 7Be. However, the 7Be concentration levels in near surface air are not expected to respond immediately to the change of elevation of the tropopause. The calculation of time lag is the next step in understanding the atmospheric procedures behind the correlation. This step will also reveal information about radioactive aerosols movement in the atmosphere's transfer cycles.

The time lag was found by holding the same column of data for 7Be and calculating the correlation coefficient (R) for each new column of daily data of the tropopause height. The new columns are created by going back in time with a step of one day waiting for the best correlation in order to find how many days we have to wait until the concentration of 7Be responds to the elevation of the tropopause height.

The correlation coefficient (R) between 7Be and the tropopause height was successively calculated for different time lags starting with lag equal to zero. The analysis of the daily data revealed that the time delay between the elevation of tropopause and 7Be concentrations in surface air is about 3days. The most striking feature of the data is the four day plateau (including the day of the measurement and the three previous days) revealing persistence in the state of the atmosphere.

In order to test our results and confirm that the correlation depends on the successive waves of air masses of a four day period a cumulative index should be calculated.

The results improved as the correlation coefficient hits a peak in the first calculation at 0.47 and 0.46 in the second one. This indicates that the time lag is 2 to 3 days as the first two calculations refer to the correlation between the 7Be concentrations and 4day average (3 previous days and the day we measured 7Be) measurements of tropopause heights. .

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Airborne radionuclides measured in Wako, Japan, after the Fukushima Dai-ichi nuclear power plant accident in 2011

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On March 11, 2011, an earthquake of magnitude 9.0 occurred near the east coast of Honshu, Japan, and was followed by a large tsunami. The disasters caused damage to the Fukushima Dai-ichi nuclear power plant (FDNPP), resulting in the release of radionuclides into the environment. Hence, we initiated an urgent measurement of the radioactivity concentrations of airborne radionuclides at the RIKEN Wako Institute (35°46'44'' N, 139°36'54'' E), Wako, Japan, located about 220 km to the southwest of FDNPP.

Air dust was collected using a commercially available air dust sampler (M Enterprise SP-30). Dust was collected for 30 min for the first two samples F01 and F02 in the period March 15 11:15–11:45 (Japan Standard Time, JST) and March 16 13:15–13:45, respectively. After the third sample from March 16 18:32 to March 17 9:00, dust was continuously collected for about one year using a total of 84 filters (F03–F86). The filter samples were subjected to γ-ray spectrometry using a Ge detector.

In this work, radioactivity concentrations were determined for Ba-140, Cs-137, Cs-136g, Cs-134g, I-133g, Te-132, I-131, Te-129m, Ag-110m, Mo-99, and Nb-95g. The highest radioactivity concentrations for typical radionuclides in unit Bq/m^3 were 8.8 ± 0.2 for Cs-137, 1.2 ± 0.2 for Cs-136g, 8.5 ± 0.2 for Cs-134g, 4.7 ± 0.3 for I-133g, 58 ± 2 for Te-132, 8.0 ± 1.2 for Te-129m, and 35 ± 1 for I-131 in the F01 sample The time variations of the radioactivity concentrations and their ratios in Wako are discussed by referring to those of the ambient effective dose rate as well as the amount of rainfall

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Coprecipitation of Radionuclide Microquantities on Chitosans of Different Molecular Masses in Solutions

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The current approaches to spent nuclear fuel treatment lead to the occurrence of radionuclides in repositories and ponds for the storage of fuel elements. In addition, radionuclides, including the above-mentioned ones, are components of low-activity liquid waste. Despite the efforts taken to localize radioactive compounds, they penetrate into the environment, including the World Ocean. The usage of chitosans and their derivatives as flocculants in sewage water purification from heavy metals, dyes, and surfactants is reported in [1, 2]. The advantages of ashless organic coprecipitators over inorganic ones are described in [3]. The authors showed that the degree of Pu extraction from 0.01 M solutions of HNO_3 on high-molecular chitosan (HMC) with MM = 15⋅105 g/mol and low-molecular chitosan (LMC) with molecular masses (MM) = 5⋅103 g/mol is 95% to 99%. Simultaneously, it was established that the coprecipitation degree of ^{60}Co и ^{54}Mn radionuclides ranged from 78% to 85%, and that of ^{90}Sr and ^{137}Cs was 25-30%, irrespective of the MM of the chitosans and the solution compositions. The data on the behavior of other radionuclides during flocculation on chitosans is unavailable in the literature.

Based on the above considerations, the aim of this study was to study the behavior of radionuclides during coprecipitation on chitosans from solutions of complex chemical compositions. The possibility of ^{233}U, ^{239}Pu, ^{241}Am, ^{152}Eu, ^{90}Sr, ^{90}Y, and ^{60}Co coprecipitation on chitosans of different MM was studied.

At first, the solubility of the obtained chitosans in different media was studied. It was shown that HMC and LMC were insoluble in distilled water and dissolved well at pH < 3. The solubility of chitosans in those solutions ranged from 8 g/l to 10 g/l and 10 g/l to 15 g/l for HMC and LMC, respectively. Increasing the solution pH to 6 for LMC and 8 for HMC resulted in the formation of a bulk thick precipitate. In both cases, the residual chitosan concentration was 0.45 g/l. In sea water at pH = 8.5, the solubility of HMC and LMC decreased to 0.045 g/l.

It was found that the efficiency of the sorption of the ^{233}U, ^{241}Am, ^{152}Eu, and ^{60}Co radionuclides on crystallized HMC and LMC from salt solutions is low, with the distribution coefficients Kd being not higher than 100 ml/g after the time of contact between the solid and liquid phases of 1 h and at V/m = 100 ml/g. Since the studied chitosans displayed a low sorptive capacity, our further research was focused on the flocculation coprecipitation of radionuclides.

As follow from the data on the ^{233}U, ^{239}Pu, ^{241}Am, ^{152}Eu, ^{90}Sr, ^{90}Y, and ^{60}Co coprecipitation on HMC and LMC in solution, in both cases degree of coprecipitation a of all radionuclides studied, except ^{60}Co and ^{90}Sr, reached virtually peak values at a chitosan concentration of 1 g/l. For HMC, a was 80% for ^{152}Eu and ^{90}Y, 99% for ^{233}U and ^{241}Am, and 85% for ^{239}Pu. In contrast to An, ^{152}Eu, and ^{90}Y, the a for ^{60}Co increased monotonically with increasing the HMC concentration in solution and at [HMC] = 5 g/l, reached 40%. For ^{90}Sr, a was not higher than 3% over the entire chitosan concentration range. For LMC a for An, ^{152}Eu, and ^{90}Y varied insignificantly ranging from 92% to 99%. For ^{60}Co and ^{90}Sr, a increased to 40% in the chitosan concentration range 0-1 g/l. Increasing the [LMC] in solution further on had hardly any impact on a for ^{90}Sr, by increased it monotonically for ^{60}Co. The obtained a values were 40% and 60% for ^{90}Sr and ^{60}Co, respectively, at [LMC] = 5 g/l. A comparison of the results showed that the coprecipitation of all the elements studied was more effective on LMC than HMC.

To conclude, using chitosans for concentrating radionuclides from salt solutions could find practical use during not only ecological monitoring of natural waters, but also the reprocessing of low-level liquid waste.

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Environmental impact due to the operation of a tin and lead industry inferred by lichens

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For the last 20 years, the brazilian most important tin and lead industry has been producing these metals in Pirapora do Bom Jesus, a city placed in state of Sao Paulo. As a consequence of the industrial process, wastes are released into the environment mainly as dust and in slag which is stored in piles in open air. The concentration of natural radioactivity can be increased as well as the trace elements by the industrial process. This paper analyzed the environmental impact due to the operation of a tin and lead industry using lichens as bioindicator. The lichen specie chosen was Canoparmelia texana because it is one of the most widely spread in natural ecosystem and also in polluted urban areas. Samples of these specie and soil were collected around the industry and the concentrations of the natural radionuclides from 238U and 232Th series, trace elements and lead isotopic ratio were determined using the techniques neutron activation analysis (NAA), alpha and gamma spectrometry, gross alpha and beta counting and thermal ionization mass spectrometer (TIMS), respectively. The lichen samples collected closer to the industry presented the highest concentrations of 238U, 226Ra, 210Pb, 232Th, 228Ra, Hf and Ta and, by the results of lead isotopic ratio, it was possible verifies the fingerprint of the contamination conforming the efficiency of lichens as bioindicator in environmental studies.

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Spectroscopic Studies of Complexation Behaviour of Uranium(VI) by Schiff Bases

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Uranium can be released into the natural environment especially from mining areas by weathering, erosion and anthropogenic activities as well as by nuclear incidents and thus represents a hazard potential for humans. New supramolecular complexing agents with N, O, S donor function are developed for the use in nuclear field and environmental protection to separate the metals of the d- and f-block and thus to clean contaminated areas. An essential basic component of these new organic ligands are Schiff bases.
In this study the complexation of uranium(VI) with Schiff bases N-benzylideneaniline (NBA), 2-(2-hydroxybenzylidenamino)phenol (HBAP) and alpha-(4-hydroxyphenylimino)-p-cresol (HPIC) was investigated in alcoholic solution using the UV-vis spectroscopy and time-resolved laser fluorescence spectroscopy with ultrashort laser pulses (fs-TRLFS). Through the change of the absorption or emission properties of organic ligands can be observed the complexation with uranium(VI) and calculated corresponding complex formation constants.
The complexation of uranium(VI) with NBA was observed by a hypsochromic shift in the NBA band to 237 nm with the UV-vis spectroscopy. Investigations with the ligand HBAP show a bathochromic shift to 281 nm. The UV-vis absorption spectra of HPIC with uranium(VI) show no spectral shift, but a decrease in intensity of the double band at 283nm and 332 nm in comparison to the free ligand. All three ligands form complexes MLx with more ligand molecules (x=2,3).
The fs-TRLFS as a sensitive speciation technique was used to determine the luminescence properties of formed complexes in the uranium(VI)-NBA, uranium(VI)-HBAP and uranium(VI)-HPIC systems. The emission signals had a hypsochromic, bathochromic and hypsochromic shift in comparison to the emission maxima of the uncomplexed ligand. This fs-TRLFS investigation opens up the possibilities for the determination of very short-lived complex species via the fluorescence of the organic compounds by delocalized π-electron systems.

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Preparation of spiked grass for use as environmental radioactivity calibration standard

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The monitoring of environmental radioactivity is important for public health protection. It is all the more important as the radionuclides can enter the food chain. Environmental nuclear analysis is usually carried out using γ-ray spectrometry on homogenized raw or treated materials (i.e. dried, sieved or ashed samples). The composition and density of environmental monitoring samples are very different from a sample to another.
The Laboratoire National Henri Becquerel (LNE-LNHB), as the French primary laboratory in the field of ionizing radiation, is working on the production of suitable calibration standards to improve the traceability chain of environmental radioactivity measurements. To address this issue, LNE-LNHB intends to produce mixed γ-ray calibration standards with certified mass activity and composition as representative of real environmental samples as possible. The use of such standards, suitable in radionuclide composition and density, will also improve the measurement system calibration due to a more accurate correction of self-attenuation by measuring a known sample whose composition is close to the real ones.
The paper describes the preparation and characterization of a low density matrix made of treated real grass spiked with mixed γ-ray emitters. The composition of the spiking cocktail includes low energy γ-ray emitters to account for self-attenuation in the matrix and high energy γ-ray emitters to cover a wide energy range. Multi- γ emitters have also been included to test the ability to estimate coincidence corrections. The spiking yield of the matrix is assessed and the homogeneity of the batch is verified by variance analysis on subsamples.
A proficiency test is being organized with the resulting matrix. The samples are to be shipped on mid-September so; unfortunately the results of the intercomparison will not be known by the time of the conference.

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Gamma external radiation dose for Mexican population

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Since 1992 the Chemistry Faculty of the National University of Mexico has been performed studies about environmental natural radioactivity, gamma, radon and thoron levels in the Metropolitan Zone of Mexico City (MZMC) and other cities like the Metropolitan Zone of Guadalajara (MZG). This work report the gamma absorbed dose rate measured at the beginning with Ca SO4: Dy + PTFE and in the last studies with a LiF; Mg, Cu, P+PTFE thermoluminiscent dosimeters developed at the National Institute of Nuclear Research, ININ. This dosimeter fulfills the ANSI-N-545 code for environmental monitoring. Each dosimetric-plastic package contains two pellets, that were placed at 1.50 to 2 m above the floor at indoor dwellings and exposed. The total sampling period was one year divided in four periods of three months each one. TLD's were evaluated in a Harshaw analyzer Model 4000 coupled to a PC. Mean arithmetic value of gamma absorbed dose rate at indoor air in the MZG dwellings (not-subtracted cosmic background) was 0.12 μGy.h−1 as mean with fluctuations from 0.07 to 0.15 μGy.h−1. The arithmetic mean value is higher than that obtained in the Metropolitan Zone of Mexico City, in spite that the MZG altitude is lesser than the first one; % RMD average precision for duplicate pairs of gamma exposure rate was lower than 5%. Taking into account the conversion factor of 0.7 Sv.Gy−1 for gamma-rays and an indoor occupancy factor of 80% the annual effective equivalent dose was calculated and compared to the to the world average.

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EC Interlaboratory Comparison on Radionuclides in Dried Bilberries

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The Euratom Treaty obliges Member States of the European Union to perform measurements of the radioactivity on their territories and to report the results to the European Commission. Therefore, regular European comparisons are conducted in order to verify the performance of the monitoring laboratories. In 2011, the Institute for Reference Materials and Measurements (IRMM) organised an interlaboratory comparison (ILC) for the activity concentrations of three radionuclides (40K, 90Sr, 137Cs) in dried bilberry powder. The Fukushima accident underlined the importance of radioactivity monitoring in food, and the need for comparisons and proficiency tests as a tool to provide reliable measurement results.

The candidate reference material IRMM-426 Wild Berries was used as a testing material. The berries were collected in the region affected by the Chernobyl accident. Due to the natural uptake from elevated levels in the environment, the radionuclides 137Cs and 90Sr were metabolised by the plants, hence no spiking was required. The material was processed at IRMM and the reference values traceable to SI units and SIR were established in the CCRI(II) supplementary comparison in which nine National Metrology Institutes participated [1].

In total, 88 monitoring laboratories participated in this ILC. They were free to use methods of their own choice, preferably the routine procedures used in their laboratories. The measurement procedures used by the participants are discussed in this paper in an overview of methods. The results of the participating laboratories were evaluated versus the reference values. A robust evaluation of the performance of individual laboratories was performed using relative deviations and En numbers [2].

The activity concentrations of 137Cs and 40K were determined almost exclusively by gamma-ray spectrometry with 9 % and 18 % of results deviating more than 20 % from the reference values of (772 +/- 32) Bq•kg-1 and (250 +/- 17) Bq•kg-1, respectively. These results are worse in comparison to previous ILCs. This may be due to the food matrix and inappropriate use of corrections for differences in density and/or geometry between sample and standard sources of activity. In the case of 90Sr, about 77 % of results lie within 20 % from the reference value of (153 +/- 8) Bq•kg-1 and 69 % of results are En compatible. This is much more favourable than observed in previous exercises.

This comparison demonstrates that several laboratories have difficulties to determine activity concentrations of 137Cs and 40K in food samples. The relative success in the 90Sr determination might be attributed to the easier separation of strontium from the dried fruit matrix compared to milk powder or soil in earlier comparisons.

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Anomalous uranium enrichment in coals from Odeř, Sokolov Basin, Czech Republic

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Contents of uranium in coals from Odeř in the northernmost part of the Sokolov Basin, Czech Republic, reach extremely high values, up to several wt%. The coal seam is situated in the vicinity of well known St. Joachimsthal uranium ore deposits. The coals from Odeř can be classified as xylitic to detritic brown coal orthophase.

The present study has been aimed at investigation of this uraniferous mineralization by detailed characterization of the anomalous coals in comparison with a representative set of Czech coals. The characterization included proximate and ultimate analyses, multielement analysis by neutron and photon activation analyses, structural characterization by infrared microspectroscopy, and micropetrographic analysis.

Contents of uranium in the studied Odeř coals ranged from 45 ppm to 6 wt%, contrary to common contents in other Czech coals not reaching a ten ppm level. The anomalous uranium levels seem to be associated with organic matter as indicated by positive correlation between uranium and oxygen (dry, ash free) contents in Odeř coals, contrary to other coals where the uranium content is correlated rather with the mineral (clay) fraction. The infrared microspectroscopy showed that the uranium enrichment was accompanied with decrease in aliphatic C-H bonds and slight increase in oxygen functional groups. The observed uranium microclusters are not formed by uranyl species, but probably by uraninite. These clusters with signs of strong thermal or radiolytic alteration of surrounding coal matter were clearly observed also by micropetrography.

The study has confirmed previously proposed explanation of uraniferous mineralization in sedimentary carboniferous substances by the mechanism of fixation and reduction of soluble uranyl species (e.g., humic, carbonate/hydroxo complexes) by sedimentary organic matter under diagenetic or hydrothermal conditions, and formation of uraninite. The process is accompanied with oxidation of alcohol functional groups and dehydrogenation of aliphatic C-H bonds, and release of molecular hydrogen and protons.

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Study of Uranium Behavior in Lignite Sediments from Ruprechtov Natural Analogue Site

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Study of uranium oxidation states in natural objects is important to estimate radioecological impact of U and as a natural redox monitor for assessing geological sites considered for long-term highly radioactive waste storage. In this work we studied U oxidation state distribution in the sediment samples by wet chemistry and synchrotron radiation method XAS. Novel wet chemical method for separating U(IV) and U(VI) from solid samples was developed and utilized. Two lignite samples having 172 and 539 ppm of U collected from the sediment layer of the Ruprechtov natural analogue site in Czech Republic were studied by described methods. Redox processes during U extraction has been monitored using 236-U(IV) and 236-U(VI) tracers. Ascorbic acid was used as selective reducing agent for Fe(III) to minimize its oxidizing effect on U(IV). XAS showed for both samples, that bulk of U in the samples is U(IV). Dissolution experiments showed that U(IV) amount varied widely resulting from the strong oxidation caused by Fe3+. Use of ascorbic acid reduced considerably the effect of Fe3+ on U redox balance: about 75% of U after samples' extraction was found in U4+. 234-U/238-U activity ratios (AR) in U(IV) and U(VI) fractions were used as an additional monitor for U redox processes taking place during materials extraction. AR in U(IV) fraction, which is less than 0.5, did not change due to extraction induced oxidation, whereas in U(VI) fraction AR was observed to decrease dramatically, depending on the amount of U(IV) oxidised during the extraction. AR for U(VI) was found below unity after 10 min extraction with pure acid solution but with addition of ascorbic acid AR increased up to 2. Results showed good agreement with lignite samples from other regions of Ruprechtov site.

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Selective liquid-liquid extraction of Sr-85 with modified calixarenes

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Sr-90 is a long-lived radionuclide (T(1/2) = 28.6 a), produced as a by-product in nuclear power plants. Due to its chemically similarity to calcium, it follows the food chain from environment (e.g. aquatic systems and soil) to fauna and human in case of release in the biosphere. Strontium can be, as well as calcium, incorporated in bones. Stable isotopes of strontium might not be harmful, but radioactive strontium can lead to bone disorders and diseases, including leukaemia[1].
Calixarenes and their functionalised derivatives are research subjects in the development of extracting agents, transporters, stationary phases[2] or bio-sensors. We used modified calixarenes, including derivatives having carbonyl binding sites, for the extraction of strontium[3] by means of a liquid-liquid extraction in a chloroform / water system. As a simulated contamination solution with Sr-90, the aqueous strontium phase was traced using the short-lived radionuclide Sr-85 (T(1/2) = 64.9 d), which was produced and purified at the in-house 18 MeV-cyclotron[4].
Under alkaline conditions, strontium extraction yields of >(90±4)% were obtained. Furthermore, the impact of inorganic and organic impurities, competing ions like sodium, calcium, acetate or tartaric acid, to the extraction performance was studied. The used carboxy-modified calixarenes are highly potent and selective extracting agents towards strontium, under conditions near to nature (e.g. synthetic groundwater).

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Poster Session / 115

CROCK: Crystalline Rock Retention Processes A 7th Framework Programme Collaborative Project (2011-2013)

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The EURATOM FP7 Collaborative Project "Crystalline Rock Retention Processes" (CP CROCK) is established with the overall objective to develop a methodology for decreasing the uncertainty in the long-term prediction of the radionuclide migration in the crystalline rock far-field. The project is launched in response to the need identified in conjunction with selection of retention data for the forthcoming crystalline host-rock HLW disposal Safety Case. The process of selecting a set of data for this purpose showed that the spread in data is broad and that this spread in data cannot presently be related to material properties or processes. Consequently, very conservative numbers need to be used in order to be defendable within the Safety Case.

The project makes use of the broad set of existing analytical approaches, methodologies, and general knowledge from decades of past investigations. It builds on the output and main conclusions of the 6th FP IP FUNMIG project and the Swedish site selection program. The experimental program reaches from the nano-resolution to the Performance Assessment (PA) relevant real site scale, delineating physical and chemical retention processes. Existing and new analytical information provided within the project is used to set up step-wise methodologies for up-scaling of processes from the nano-scale through to the PA relevant km-scale. Modeling includes testing up-scaling process and parameters for the application to PA and in particular, the reduction of uncertainty.

The project started on 1st January 2011 and will last 2 years and a half (2011-2013).The project is implemented by a consortium with 10 Beneficiaries consisting of large European Research Institutions, Universities and SME's and from countries with dedicated crystalline host-rock disposal programs and particular competence in this field. National Waste Management organizations participate as associated groups, contributing with co-funding to beneficiaries, infrastructure, knowledge and information. They also contribute together with national regulators to guidance with respect to application of the project to the disposal Safety Case and scientific-technical review.

The research leading to these results has received funding from the European Atomic Energy Community's Seventh Framework Programme (FP7/2007-2011) under grant agreement No. 269658 (CROCK).

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Iodine-129 and iodine-127 in soils from Germany

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The environmental abundance of 129I has been changed substantially, mainly as a consequence of the 129I releases from European reprocessing plants [1, 2]. Iodine from wet and dry deposition is accumulated in soils, transported by surface waters, infiltrates groundwater, and makes its way through the biosphere. One of the goals of this project is to investigate the inventories of 129I and 127I in the pedosphere in Germany.
Stable iodine is analysed by using inductivity coupled plasma mass spectrometry (ICP-MS). The 129I /127I ratio is determined by means of accelerator mass spectrometry (AMS).
Sampling of different soil types at various locations in Germany, down to a depth of 50 cm, is in progress. The samples are taken from 0-5 cm, 5-10 cm, 10-20 cm, 20-30 cm, and 30-50 cm, resp.
The 129I inventories of the first soils analysed range from 120 mBq m-2 to 470 mBq m-2 (depth: 50 cm), with the higher values being found in the northern and western parts of Germany. The profiles show a characteristic distribution of the 129I /127I ratios, with the highest values in the organic rich topsoil layer and a distinctive decrease of the values with increasing depth. This indicates that anthropogenic 129I is strongly associated to soil organic matter, and is released again in a very slow process.

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Sorption of niobium on Olkiluoto soil samples

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A KBS-3-type repository for the spent fuel from the Finnish nuclear power reactors in Olkiluoto and Loviisa is to be built in the bedrock at the Olkiluoto site at the depth of approximately 400 m. The final disposal plan includes a safety assessment of the spent nuclear fuel, where the potential dose contributing nuclear waste nuclides for man are specified. As a part of this assessment, the transport of these prioritized radionuclides from the geosphere to surface environment and their fate within is modelled and evaluated. Nb-94 is classified as high priority radionuclide in the long-term safety assessment of spent nuclear fuel.

The retention of niobium was studied on Olkiluoto soil samples representing humus layer and mineral soil layers. Sampling extended from the soil surface to the bedrock surface. Soil samples were used without pretreatment, e.g. drying or sieving. Mass distribution coefficient, Kd, describing the effectiveness of the retention was determined by batch sorption tests. The liquid phase used in the tests was synthetic soil solution simulant, which composition is similar to the composition of Olkiluoto soil solution. The equilibrium time ranged from one day to three weeks for humus samples and from one week to nine weeks for mineral soil samples. The final activity of the Nb-95 tracer was determined by gammaspectrometry with Wizard ™ 3''.

The sorption of niobium was found to be high on mineral soil samples as the Kd values ranged between 1.1x10^3 ml/g and 1.0x10^6 ml/g. Niobium was retained on colloidal particles which was seen as an increase in the Kd values upon filtering. Sorption showed no clear dependence on time or sample depth. The sorption on humus samples was smaller as the Kd values ranged from 230 ml/g to 1.2x10^3 ml/g.

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TiO2 based absorber for uranium separation and 236U measurement with AMS

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The uranium separation from natural waters was studied at the Department of Nuclear Chemistry in late 80´s and early 90's. TiO-PAN absorber (titanium dioxide embedded in polyacrylonitrile) showed high sorption capacity for this element.

This composite absorber is planned to be used for uranium pre-concentration for measuring environmental 236U/U ratios by Accelerator Mass Spectrometry (AMS). After the preliminary AMS results showing anthropogenic contamination in TiO-PAN absorber, the preparation of titanium oxide was studied with a strong emphasis on elimination of this contamination.

Hence, titanium dioxide materials were prepared from an organic uranium-free compound. These samples were then characterized for their crystal structure, their specific surface area, and their sorption towards uranium. Detailed sorption properties were determined also for TiO-PAN absorber including kinetic experiments, sorption isotherm, and effect of several ions (Ca^{2+}; Mg^{2+}; Fe^{3+}; NO_3^-; SO_4^{2-}; Cl^-; CO_3^{2-}). In these experiments, the uranium concentrations were measured by Time Resolved Laser Induces Fluorescence Spectrometry (TRLFS), which allows us to measure uranium concentrations up to ppb level. A strong influence of Cl- ions present in the solutions was seen, thus the calibration of TRLFS has to be made in order to determine the correct uranium concentration.

Presently we are focusing on finding a water source which is not anthropogenically contaminated in order to use this water for further experiments and on the following separation and concentration steps for AMS sample preparation.

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Fractionation of U, Th, Ra and Pb from boreal forest soils by sequential extractions

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To study the mobility of the natural radionuclides in forest soil a five step sequential extraction procedure was carried out to soil samples taken from various depths down to three meters at the Olkiluoto Island, Finland, where the final disposal repository of spent nuclear fuel is planned to be constructed in the bedrock. The studied extraction fractions were exchangeable, acid-soluble, reducible, oxidizable and tightly bound. It was found that the extractability of most of the studied radionuclides was dependent on the sample grain size and depth. All the elements were found to be concentrated in the smallest grain size samples (<0.063mm). The extraction behaviour of thorium, however, did not vary with sample depth and only about 10 % of thorium extracted until the final extraction step. Stable lead and Pb-210, as well as barium and radium concentrations were found to correlate strongly in the extractions. Radium and barium were leached more readily than the other elements, approximately 17 % of total radium was found in the first extraction fraction representing exchangeable ions. Uranium was found to be more mobile in the top-soil horizons compared to the horizons further down. In the top-soil samples an average of 51% of extractable uranium was already leached in the second extraction step representing the elements soluble in weak acids, whereas only 13% of the uranium in the sub-soil samples was extracted in this step. This might be due to the changes in the soil redox-conditions while going further down in the soil profile. Also the extraction behaviour of lead and iron might suggest more reducing conditions in deeper soil horizons because the extractable percentage of lead and iron in oxidizable-fraction increased with sample depth.

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Redox behavior of the Tc(VII)/Tc(IV) couple in dilute to concentrated NaCl and MgCl2 solutions

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For the long-term performance assessment of nuclear waste repositories, reliable predictions of radionuclide mobility have utmost importance. 99Tc is a β-emitting long-lived (T½ ~ 211.000 a) fission product highly relevant for nuclear waste disposal. Tc(VII) and Tc(IV) are the most stable and dominant redox states of technetium under environmental conditions. Heptavalent Tc exists under non-reducing conditions as the highly soluble TcO_4^- pertechnetate anion. Technetium present in the tetravalent oxidation state, however, forms hydrous oxides ($TcO_2 \cdot xH_2O(s)$) which drastically limit Tc solubility and mobility. Because of the significant impact on Tc chemistry, the redox behaviour of the Tc(VII)/Tc(IV) couple needs to be investigated in detail. In order to predict Tc behaviour for nuclear waste disposal in rock salt formations, dedicated studies under saline conditions are required.

In this study, the redox behaviour of the Tc(VII)/Tc(IV) couple was investigated in dilute to concentrated saline systems. Reduction experiments were performed under Ar atmosphere by using different homogenous and heterogeneous reducing systems in NaCl (0.5 M and 5.0 M) and $MgCl_2$ (0.25 M, 2.0 M and 4.5 M) solutions to provide for repository-relevant geochemical conditions. Eh and pH were measured after given aging times and the results systematised according to Pourbaix diagrams in order to assess the redox behaviour of technetium in highly saline systems. Technetium concentrations, redox state and aqueous speciation were analysed. The results show a clear effect of the specific reducing systems and ionic strength conditions on measured Eh values and Tc(VII)/Tc(IV) redox transformations. The experimental data are compared to thermodynamic calculations and geochemical model predictions.

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Pu-240/Pu-239 atom ratios in the northern North Pacific and equatorial Pacific water columns

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Anthropogenic radionuclides such as Pu-239 (half-life: 24,110 years), Pu-240 (half-life: 6,564 years) and Pu-241 (half-life: 14.35 years) have been released into the environment as the result of atmospheric nuclear weapons testing, disposal of nuclear wastes and nuclear fuel-cycle reprocessing operations, etc. In the North Pacific Ocean, two distinct sources of Pu isotopes can be identified; i.e., the global stratospheric fallout and close-in tropospheric fallout from nuclear weapons testing at the Pacific Proving Grounds (PPG) in the Marshall Islands. The atom ratio of Pu-240/Pu-239 is a powerful fingerprint to identify the sources of Pu in the ocean.
The objectives of this study are to measure the Pu-240/Pu-239 atom ratios in seawater from the northern North Pacific Ocean and the equatorial Pacific Ocean and to discuss the transport processes of Pu.
The Pu-240/Pu-239 atom ratios were measured with a double-focusing SF-ICP-MS, which was equipped with a guard electrode to eliminate secondary discharge in the plasma and to enhance overall sensitivity.
In the equatorial Pacific, Pu-240/Pu-239 atom ratios were 0.215 in the surface water and increased gradually with depth reaching 0.267 at the 3000 m depth after which they decreased with depth to 0.228 at the bottom layer. In the northern North Pacific, Pu-240/Pu-239 atom ratios showed no notable variation from subsurface water of 100 m depth to deep water of 2000 m depth, then increased with depth to 0.255 at the bottom layer. The atom ratios in water columns of the northern North Pacific and equatorial Pacific were significantly higher than the mean global fallout ratio of 0.18. High atom ratios of Pu-240/Pu-239 prove the presence of close-in tropospheric fallout from nuclear weapons testing at the Pacific Proving Grounds.

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Improvement of Detection Limits for Gamma-Ray Emitting Naturally Occurring Radionuclides in Drinking Water and Biological Materials by Instrumental Analysis using Compton Suppression Spectrometry

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The instrumental measurement of very low activities from naturally occurring radioactive materials (NORM) by gamma-ray spectrometry requires the use of detection systems with the lowest possible external activity. Generally, lead shielding and special detector construction materials are used for this purpose. In our laboratory, we have combined a lead shield and a Compton suppression system (CSS) to lower the external background activity. The external background was evaluated by first counting the empty detector for 24 h without the lead shielding in conventional mode (A). Then the same procedure was repeated with the lead shield (B). A third counting was done using the CSS and the Pb shield (C). A background reduction factor of approximately 3 was obtained by comparing the spectra collected using systems (A) and (B). When the conventional and the CSS with Pb shielding (i.e. B and C) were compared, the average background reduction factor was about 16. It was then possible to identify many radionuclides in tap water, well water, spring water, mussel tissues and oyster tissues. For example, the following radionuclides were detected in a canned tuna sample: (i) 234U, 234Th, 230Th, 214Pb, and 214Bi belonging to the 238U decay series; (ii) 235U, 231Th, 231Pa, and 227Th of 235U decay series; and (iii) 228Ac, 228Th, 212Pb, and 214Bi of the 232Th decay series. A comparison of the minimum detectable activity (MDA) values showed that the CSS gave the lowest value for 228Ac, 212Pb, 212Bi and 208Tl while comparable values were obtained for other radionuclides.

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Atmospheric activity concentration of radiocesium at Mikamine, Sendai and radioactivity distribution on the collection filters used in the measurement.

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The Fukushima 1 Nuclear Power Plant suffered major damage from the 2011 off the Pacific coast of Tohoku Earthquake and subsequent tsunami on March 11, 2011 and released various radionuclides. Monitoring of environmental radioactivity should provide important information on the behavior of the radionuclides.
To investigate the time variation of atmospheric activity concentration of radiocesium, we have regularly collected aerosol particle samples at Mikamine, Sendai from March 15, 2011 to present. The aerosol particle was collected on a cellulose-glass fiber (Advantec, HE-40T). The radioactivity of radiocesium in the aerosols collected was determined by gamma-ray spectrometry using HP-Ge detectors. The atmospheric activity concentrations of radiocesium were calculated as the ratio between the radiocesium activity in the aerosol particle sample and the total volume of sampling air. As a result, it was found that the maximum activity concentrations of radiocesium were recorded in March 20-21, 2011 and then the atmospheric activity concentrations of radiocesium roughly decreased with a half-life of 10 - 30 d.
In addition, radioactivity distribution on the collection filters used in the measurement was measured by using imaging plate technique. The relation between the monitoring results on the concentration of radiocesium in the air and the imaging plate results are discussed in the presentation.

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Radioactivity measurement for air-dust and soil collected in eastern Japan area after the nuclear accident at the Fukushima Daiichi Nuclear Power Station

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On March 12, 2011, a large amount of radioactive nuclides have been released into the environment by the nuclear accident at the Fukushima Daiichi Nuclear Power Station .
Our group started radioactivity measurements for I-131, Cs-134, Cs-137 and the other radio nuclides from air dust collected using high-volume air sampler by germanium semiconductor detector soon after the accident. Air dust samples have been collected at some spots in eastern japan periodically. In some cases, we have used charcoal filter to collect I-131 in the form of gas, and collections of air dust by cascade impactor were also performed to separate air dust according to molecular mass. We have already measured more than 500 air dust samples, and we will discuss the distribution and time variation of radionuclides in the environment.
In addition to gamma-ray measurement targeting I-131, Cs-134 and Cs-137, our group has been carrying out detection of pure beta emitter like Sr-89 and Sr-90 in soil and filter using chemical separation technique. Because Sr-89 is a short life radionuclide (t1/2 = 50.5 d), the existence of Sr-89 represents the radionuclide is originated from the accident. For this purpose, we developed strontium isolation technique using solid-phase extraction method and selective detection technique of radioactivity of Sr-89 and Sr-90 using Cerenkov counting.

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The Thermal Decomposition of CH3131I in a Gas Phase

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The localization of volatile radioactive iodine compounds by various sorbents from vapor-gas media is a vital issue for environmental protection during both irradiated nuclear fuel reprocessing and accidents at nuclear power enterprises, including nuclear power plants (NPPs).

The thermal decomposition of methyl iodide CH3131I, a volatile radioactive iodine organic compound, in a gas flow in the presence of various modifications of "Fizkhmin"TM granulated materials based on silica gel impregnated with d-elements were studied.

Test facility included the following basic parts: rotameters; a CH3131I generator; scrubber with water; the heating furnace of mine type; composite materials under study; the thermocouple; a column with SiO2-Cuo; scrubber with 0.05 M Na2SO3 solution; the heating furnace of tubular type; columns with SiO2-AgNO3.

It was found that in the absence of "Fizkhmin"TM material, the degree of the decomposition of CH3131I (10 mg) in air (flow rate 4.5–5.5 cm/s and time of the air flow presence in the heating zone 1.0 – 1.5 s) was equal to ≈(7-10)% at (540 ± 10)oC, ~(70-75)% at (640 ± 10)oC, and ~(97-99)% at (770 ± 15)oC.

In the presence of silica gel granules measuring 1.0-3.0 mm, the degree of the decomposition of CH3131I (10 mg) in air (linear flow rate 4.8–5.2 cm/s and time of the air flow presence in the heating zone 1.0–1.1 s) was equal to ≈(2-3)% at (240 ± 10)oC, ~(10-15)% at (340 ± 10)oC, ~(75-80)% at (440 ± 10)oC, and ~(97-99)% at ~(540 ± 10)oC. Silica gel granules allow decreasing the CH3131I thermal decomposition temperature in an air flow by ~200oC.

In the presence of "Fizkhmin"TM granulated materials impregnated with Ni compounds or Ni-Cu mixture (8-10 wt.% and granule size 1.0–3.0 mm), the degree of the decomposition of CH3131I (10 mg) in air (linear flow rate 4.8–5.2 cm/s and time of the air flow presence in the heating zone 0.8–1.1 s) was equal to <0.2% at (20 ± 3)oC, ~(0.3-1.0)% at (150 ± 20)oC, ≈(15-30)% at (250 ± 10)oC, ~(85-92)% at (340 ± 15)oC, and ~(95-99)% at (465 ± 20)oC. "Fizkhmin"TM granules containing 8-10 wt.% Ni or its mixture with Cu, allow decreasing the CH3131I thermal decomposition temperature in an air flow by more than ~300oC.

The dependence of the CH3131I thermal decomposition degree on the concentration of a d-element in the "Fizkhmin"TM material and its storage time, as well as on the amount of CH3131I in an air flow were studied.

In conclusion, it is necessary to note, that the using of granulated composite material "Fizkhmin"TM allow to convert the more difficult localize organic form of radioactive iodine to well-localized molecular form of radioactive iodine.

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Retardation behavior of Sr and Cs in Crushed and Intact Rocks—Two potential LLW repository Taiwan host rocks

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This study investigates sorption and diffusion of Strontium (Sr) and Cesium (Cs) in two potential host rocks (granite from Kinmen Island and basalt from Penghu Island) by using batch and through-diffusion methods in order to establish a reliable safety assessment methodology. These methods were applied to crushed and intact rock samples to investigate the actual geological environment. According to solid-phase analysis, including X-ray diffraction, elemental analysis, auto radiography, and polar microscopy, the sorption component primarily contained iron–magnesium (Fe–Mg) minerals in basalt and granite. Moreover, the distribution coefficient (Kd) of Sr and Cs in various concentrations (~10−2–10−7 M) obtained from batch tests indicated a higher sorption capacity in basalt than that in granite because of the 10% Fe–Mg mineral content. The diffusion of Sr and Cs both in crushed granite and basalt reach steady state after 110 days and apparent diffusion coefficient (Da) were 3.29×10-11 m2/s (for Sr in crushed granite), 4.17×10-12 m2/s (for Sr in crushed basalt), 2.86×10-11 m2/s (for Cs in crushed granite), 1.82×10-12 m2/s (for Cs in crushed basalt), respectively. However, diffusive result (Da) of Sr and Cs in intact rocks was estimated a lower value than those obtained using crushed rocks. According to the diffusive results in crushed and intact rocks, it showed that major retardation of Sr and Cs depended on the microporous structure of tested media, such as decreases of constrictivity (δ) and increases of tortuosity(τ). In fact, the solid/liquid (S/L) ratio decreased as is the case when switching from batch to column experiments and the sorption effect on minerals became even more negligible in retardation of radionuclide migration.

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Selected Elements content in Paraguayan Wheat

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The two fold purpose of this paper is to determine the composition of selected elements in Paraguayan wheat and flour as well as to analyse the implications of the bromine/bromate content on bakery products.Despite of its importance, there is a lack of information in regard to the normal values of the concentration of minor and trace elements in wheat from Paraguay East Region or Eastern Paraguay (1). According to that, in this work, selected minor and trace elements in eight varieties of wheat from the center and south areas of Eastern Paraguay were analyzed by XRF techniques using an Isotopic Cd Source and a XR Mo tube. The analyzed elements were K, Ca, Ti, Mn Fe Cu Zn Br Rb Sr. The materials were: whole samples, flour, bran and ashes of soft wheat, Triticum Aestivum subspecie Vulgare obtained by successive milling and sieving. Soils in the Center are Rhodic Paleudult from sandstones associated with Mollic Paleudult as well as a small portion of LithicUpdisament. In the south (Itapua) dominate two type of soils: Rhodic Paleudult from basalts and Typric Kandiudox also from basalts and some sandstone. The results (mean values) of whole samples are as follow:
K 1426.7±118.6 ; Ca 213.0±30.0 ; Ti 7.7±0.6 ; Mn 41±3.1 ; Fe 39.1±6.6 ; Cu 5.9±1.0; Zn 33.0±3.0 ; Br 7.5±0.7 ; Rb 4.3±0.4 ; Sr 3.1±0.4.
The first fraction of flour which came from the center of the grain were obtained after the first milling and sieving ; next grinding and sieving yield another fraction; after the third, the fraction was mainly bran. The results were in good agreement with those from whole samples. In addition XRF measurements made on ashes from the grains were also coherent which the above.
Essential microelements like Mn Fe Cu Zn values as well as of the other analytes found here are also into the range of those from several studies made elsewhere (2).
Another element of interest is bromine specially as $KBrO_3$. The use of this compound as an additive in the bakery dough to improve the whiteness and other characteristics of bread is well known. When using second class flour, this is clearly a deceit (3). Although there are some simple methods to look for the adulterant in the row dough, no simple procedure is available to detect it in the final product. This is overcome using XRF, which is simple and direct, provides the “normal” bromine content of national or regional flour used in the bakery is known: this is the “base line”. A significant excess in the product means adulteration.

In this regard, samples of first quality commercial flours from Paraguay (N=25) and from Argentina (N=5) were also analyzed for Br and its content was found to be within the normal values of bromine in Paraguayan wheat and flours. However when analyzing bakery products, bread (N=20) and galletas (N=6) it was found that ~35% exceed the normal Br values showing bromate malpractice.

From dietary point of view it should be emphasized that $KBrO_3$ is a complete carcinogen and classified as showing clear evidence of carcinogenety (4) ,inducing renal cell tumors as well other toxic effects. Its use as food additive has been banned(3,5). In regard to Br- product of $KBrO_3$ decomposition, after new evidences of its toxicological effects at low dosis, a provisional ADI for bromide ion of 0,4mg/kg bw has been calculated and proposed(6). Therefore it is concluded that the intake due to malpraticed bakery products together with the bromide ion of total diet, easily exceeds the recommended DI. Morover the XRF technique described herein is simple, rapid and reliable.

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Research Alliance for Validation of PGAA Actinide Nuclear Data

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A Memorandum of Understanding for close collaboration in the field of prompt gamma neutron activation analysis (PGAA) has been formulated and signed by several institutions. This research alliance includes at the moment FZJ and FRM II from Germany, BNC from Hungary, and LBNL, and will be extended to LLNL and NIST from USA as well as JAERI from Japan. Besides development of PGAA and their application in various fields the generation and validation of nuclear data for actinides is in focus to update neutron capture cross sections, energies, and intensities for prompt and delayed gamma ray data. These data are important input e.g. for simulation of GENIV reactor neutronics, nuclear waste management, safeguards applications and clearance and decommissioning of nuclear installations.

5 to 10 mg actinide-oxide powder was encapsulated in aluminum or quartz and exposed to a directed cold neutron beam at the research reactor in Budapest and at FRM II, Garching. Optimization of the sample preparation as well as irradiation conditions led to an improvement in overall uncertainty and accuracy. Experimental data were corrected for neutron self-absorption and gamma attenuation in the sample and container. Revised nuclear data will be incorporated in existing nuclear data tables and compilations.

Progress of experimental work will be reported and discussed.

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OPENING LECTURE - Prompt Gamma Activation Analysis using High-Flux Cold Neutron Beam

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The Forschungsneutronenquelle Heinz Maier-Leibnitz at Garching (FRM II) is one of the largest neutron research centers in the world with almost 30 high-end instruments. The Prompt Gamma Activation Analysis (PGAA) facility is located at the cold neutron beam with a flux of 6 x 10^10 cm-2 s-1, the highest beam flux reported. The instrument has been reconstructed recently to enable elemental analyses and irradiations with this strong beam, too. High-flux PGAA made possible new applications, but also introduced brand new challenges, which are discussed in the presentation.

The contribution of the background in the prompt gamma spectrum reduced significantly thanks to reconstruction of the shielding of the instrument. The excellent signal-to-background ratio allows the measurement of samples with the masses of less than a milligram, which is practically impossible in weaker beams. For this purpose, it was necessary to find significantly lighter materials for sample packing. The reconstruction of the sample holder from less massive components is also planned to further reduce the spectral background. The use of smaller target quantities also makes possible the efficient measurement of radioactive materials and the counting of their signals above the elevated baseline from to the original activity.

The new PGAA instrument has been used for high-flux irradiations and elemental analyses of a series of different samples successfully. Air filters with pollution contents as low as a milligram or less were analyzed, and more than 10 components were determined with fair uncertainties. The most sensitive elements (B and certain rare-earths) were found to be in amounts of less than 100 pg. The elemental compositions of micro-meteorites with masses of 300–1000 μg were determined, and they were classified based on the results. In the frame of a large international collaboration, nuclear data for transuranium actinides have been remeasured. The plans of further improvements and other possible utilizations of the high-flux PGAA facility will also be discussed.

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INVITED LECTURE - Neutron activation analysis: a consolidated analytical tool in the sugarcane agroindustry

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Currently, Brazil has become a leader in development of science and technology for the agribusiness sector of sugarcane. The country has maintained a tradition of decades as the world's largest producer of sugarcane preferentially oriented to the production of sugar and ethanol. In addition, bioelectricity generated by the burning of bagasse is employed to power mills with electric power remaining being sold to make a clean process, which avoids the use of fossil fuels. More recently, innovative products derived from sugarcane as biodegradable plastics and essential amino acids have emerged and gained competitiveness in the market for economic and environmental reasons. Ethanol from sugar cane has been the most successful biofuel in the world, evidencing the enormous contribution of sugarcane to sustainability in the production of clean fuels and reduction of greenhouse gas emissions. Nevertheless, there is potential to further increase the competitiveness of products derived from sugarcane, with more sophisticated management of the production process, including quality control, optimization of partnerships between growers and industry, increased efficiency in the fermentation process, among others. To comply with the demand for one billion tons of sugarcane in 2020, new varieties with higher productivity have been developed, including genetic modified ones. For evaluating the genetic improvement, specific certified reference materials are being developed at CENA/USP. Neutron activation analysis has been successfully applied since 1986 to the various segments of the sugarcane productive chain, thereby contributing to the efficacy of the system. By assessing the amount of soil adhered to sugarcane stalks transported to the industry, tracking those mineral impurities in the entire production process, comprising washing efficiency, bagasse, juice, sugar and yeasts, it was possible to accurately measure the influence of soil type, moisture content, soil preparation techniques and the overall impact on the sugarcane payment system.

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INVITED LECTURE - Neutron Activation Analysis and Reference Materials – Development and Perfection

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This review of a 40-year practice in neutron activation analysis (NAA) is intended to illustrate the mutual benefits drawn from the values of a unique analytical tool and the enabling properties provided with the reference materials. During this period NAA transitioned rapidly to instrumental multi-element procedures based on high resolution gamma-ray spectrometry with germanium detectors. On the side of the reference materials substantial development of biological and environmental SRMs by the National Institute of Standards and Technology (NIST) (then the National Bureau of Standards) had begun in the early 1970s. Starting with the production and certification of the first ever, natural matrix, multi-element botanical SRM 1571 Orchard Leaves, issued in 1971, and followed by SRM 1577 Bovine Liver in 1972 the determination of key biological and contaminant elements, including Ca, Mg, Fe, Cu, Zn, Se, Cr, Cd, As, Pb, and Hg could be validated. The certification had been accomplished using a broad array of analytical methods, with NAA having contributed to the majority of certified values. The need for more diverse and extensively characterized SRMs to fulfill the ever-growing demands in analytical chemistry for life science research and applications has been the driving force for refinement and expansion of the NAA procedures. The development of more sophisticated procedures with detection limits down to 0.1 µg/kg enabled the recent establishment of new materials as benchmarks for current analytical investigations. NAA procedures provided critical analytical data in several recent SRM developments such as 1577c Bovine Liver, where Ag, As, Cr, and V were determined at several to tens of µg/kg levels. NAA's performance as a primary method of measurement contributed to the certification of arsenic in SRM 955c Toxic Metals in Caprine Blood, SRM 2668 Toxic Elements in Frozen Human Urine, SRM 3262 St. John's Wort Aerial Parts, SRM 3532 Calcium Dietary Supplement, SRM 3280 Multivitamin/Multielement Tablets, SRM 3233 Fortified Breakfast Cereal, SRM 1845a Whole Egg Powder, and SRM 2383a Baby Food Composite.

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INVITED LECTURE - Error, Uncertainty, and Metrology in Nuclear Analytical Methods

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Nuclear methods have well-established advantages in chemical analysis. Paradoxically, many of these advantages stem from the absence of chemistry in the analytical process. Nuclear reactions are often easier to understand than chemical reactions, and nuclear methods are usually direct, with simple equations relating the laboratory measurements to the sought-for composition. Neutron activation analysis has recently been recognized by the Comité Consultatif pour la Quantité de Matière as a primary ratio method, meaning that its results can be traceable to the fundamental units of the Système International. For this to be practically as well as formally true, an exhaustive search for, and quantification of, sources of bias (errors) and random uncertainty is necessary to describe the metrological traceability of a measurement result. The transparent interweaving of these issues with economics and fitness for purpose is an integral part of analytical research.

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INVITED LECTURE - Studies of Trace Element Species in Macromolecules and Protein Nanoclusters by Nuclear and X-Ray Techniques

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Trace elements play an important role in biological activities of living matter. A considerable amount of data on the total trace element levels in various biological tissues already exists in the literature. However, trace elements are mainly incorporated in proteins in these systems. It is important to separate these protein-bound trace elements for the identification as well as characterization of the chemical species in order to develop a mechanism and an understanding of their biological activities. We have used analytical and bioanalytical techniques such as dialysis, pH variation, ammonium sulphate precipitation, chromatofocusing, isoelectrofocusing and isotachophoresis, ion-exchange, hydroxyl apatite, size-exclusion, gas and liquid chromatography complemented by NAA, MS, and NMR to study macro-molecular species of As, Br, Ca, Cl, Cu, Fe, I, K, Mg, Mn, Mo, Na, Rb, S, Se, V, and Zn in bovine kidneys. Lately we are interested in a better understanding of the interaction of metal nanoparticles (NPs) with proteins through speciation analysis. Because of the size, some metal NPs can have significant effects on their quantum electronic and chemical properties. We are studying bio-functionalization of Au, Ag and a few other metal NPs on surfaces such as Ti that can be used for more efficient drug delivery and implant surface modification using SEM and element-specific X-ray techniques such as EXAFS, XANES and XPS.

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ORAL PRESENTATION - Analysis of Radioactive Waste Waters and Sludges int he Hungarian VVER NPP Paks

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There are some tanks at the nuclear power plant in Paks, Hungary containing sludge type radioactive waste containing more or less liquid phase too. The general physical and chemical charactersitics (density, pH, total solid, dissolved solid etc.) and chemical and radiochemical composition of these sludges are important information for volume reduction and solidification treatment of theese wastes. Based on the literature sources we have investigated and constructed a complex analysis system for the radioactive sludge and supernatant analysis, including the physical, as well as the chemical and radiochemical analysis methods. Using well known analysis techniques as ion chromatography, ICP-MS, AAS, gamma-and alfa-spectrometry and chemical alkaline fusion digestion and acidic dissolution methods we could analyze the main inorganic, organic and radioactive components of the sludges and supernatants. Determination of the mass and charge balance for the sludge samples were more difficult then for the supernantant samples. Not only are there assumptions required about the chemical form and the oxidation state of the species present in the sludge, but many of the compounds in the sludge are mixed oxides which are not directly measured. Also, the sludge is actually a slurry with a high water content. The interstitial liquid is in close contact with the sludge, and there are many ionic solubility equilibriums. The anion data for the sludge samples are based on the water soluble anions that would be available to a water wash. The water wash would not account for the insoluble hydroxides, carbonates, and mixed oxides present. The insoluble species do not contribute to the charge balance, and the cation charge is not used in the calculation. Most of the nitrate reported for the sludge is due to the interstitial liquid. Considering the limitations of these calculations, the mass balance was within the analytical error (±20%) for the sludge samples. There were three sample preparation methods used to investigate the total anion content of the sludge samples, which included water leach, potassium-hydroxide and/or sodium peroxide/sodium hydroxide fusion and acidic dissolution.

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ORAL PRESENTATION - Nuclear Forensics: age determination by the 231Pa/235U ratio

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The increase of traffics of illegal nuclear materials requires the improvement in nuclear forensics. The age of the sample, which corresponds to the determination of the date of the last purification, can give information about its origin. The aim of this work is to develop an efficient procedure which allows the determination of the age of micro-quantities. Dating micro-quantities of uranium is possible with two daughter/parent nuclide ratios: 230Th/234U and 231Pa/235U. The procedure of age determination using the 230Th/234U ratio is already effective [1,2]. The development of the 231Pa/235U dating procedure will allow to confirm results obtained with 230Th/234U. In the same time, it will provide a survey of the 235U isotope, i.e. of the enrichment rate.
In order to measure such low quantities, mass spectrometric detection was chosen. Thus, the isotopic dilution method will be used with the aim of determining the quantity of each isotope. 233Pa is the appropriate tracer for the detection of 231Pa. Therefore, 233Pa was milked from 237Np, fixed on AG1-X8 resin.
A procedure of separation has been established in order to date uranium. However, experimentally, the separation of protactinium from uranium was not effective. Consequently, distribution coefficients of protactinium have to be determined for different media (HCl and HNO3) and different type of resins (AG1-X8, TEVA, U-TEVA, TRU-SPEC) in order to optimize the procedure.
After the determination of the distribution coefficients, the different elements, i.e. protactinium and uranium, will be separated from each other without any residual amount present in each fraction.

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ORAL PRESENTATION - Prompt Gamma Activation Analysis close to Detection Limits

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The ultimate detection limits (DL) of the Prompt Gamma Activation Analysis (PGAA) method depend strongly on the amount and matrix of the measured sample as well as on the background signal contribution to the signal coming from the sample itself.

Recently, we have performed few experiments close to the detection limits for given elements at the high-flux-PGAA facility at FRM II in Garching. One of the most important element which can be determined by the PGAA technique is Hydrogen. Hydrogen impurities in Silicon as well as Hydrogen implantation in Beryllium crystals were measured and analysed. Concerning Silicon, which appears frequently in the nature and so in the PGAA samples, we could determine 3 ppm Hydrogen in the crystals. However, the constrain appeared to be the Hydrogen from the surrounding materials (which cannot be avoided) and we estimate its signal being comparable to a concentration of about 1 ppm.
The Hydrogen implantation to Beryllium crystal was measured by slightly different conditions to lower the Hydrogen contribution from the surroundings of the sample. The analysis is still in progress and exact values will be known soon.

Another experiment testing the limits of the PGAA facility at FRM II was performed in a frame of a Transmutation detectors (TMD) proposal. TMD's are small high-purity metallic foils or single crystal samples, irradiated close to the reactor core at any research or power reactor for a long time (even months). To calculate the neutron flux as well as fluence at the position, not the activity of the detectors after long-term irradiation is analysed, but the concentration of transmuted isotopes with appropriate neutron capture cross-section. The main advantage of this method is the independence of the time parameter: the information is "stamped" to the samples and can be read any time later. The samples irradiated for 21 days at the LWR-15 in Rez, Czech Republic, were pure natural Copper, Aluminum and Gold, the searched isotopes were 59-Ni and 63-Ni in Ni foil, 64-Ni in Cu foil, 198-Hg and 199-Hg in Au foil [ref.1]
In this presentation, the experiments and the results will be discussed and possible ways how to improve the detection limits will be proposed.

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ORAL PRESENTATION - Enhancing the dynamic range for high boron concentrations in low neutron capture cross-section matrices with Prompt Gamma Activation Analysis

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At the beam line of Prompt Gamma Activation Analysis (PGAA), one of the most powerful cold neutron beams in the world is available for experiments with neutrons. It offers unique possibilities to improve the detection limits, counting statistics and allows even the detection of trace elements and extremely small samples.

Problems arise, if relatively high amounts of high-cross-section elements like boron are to be detected in a matrix of elements with relatively low neutron capture cross-sections such as aluminum or silicon. In these cases the high-purity germanium detector (HPGe) can be saturated even when using small sample masses. For the case that the sample mass or the neutron flux is reduced to lower the count rates, the dynamic ranges of the peak areas do not change. The idea was to suppress the low energy gamma radiation mainly from boron more than the higher energy one from matrix elements using Pb sheets with different thicknesses. The efficiencies without lead attenuator, with 5mm and 10mm of lead in front of the detector were determined and samples with different nominal compositions made of milled powder mixtures of SiO_2, Al_2O_3, and H_3BO_3 were analyzed using the appropriate efficiencies.

The technical details of the method and the results of the first experiments will be presented.

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ORAL PRESENTATION - Comparison of Quantitative Neutron Capture Radiography, Inductively Coupled Plasma Mass Spectrometry, and Prompt Gamma Activation Analysis for Boron Determination in Biological Samples

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Category: Radioanalytical Chemistry and Nuclear Analytical Techniques
Type of presentation: Oral presentation

Comparison of Quantitative Neutron Capture Radiography, Inductively Coupled Plasma Mass Spectrometry, and
Prompt Gamma Activation Analysis for Boron Determination in Biological Samples

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G. Otto, T. Schmitz, G. Hampel

Introduction:
Boron determination is most commonly carried out in water and geological samples. Its determination in blood and tissue samples is a crucial task especially for treatment planning, preclinical research, and clinical application of BNCT. However, comparison of clinical findings remains difficult due to a variety of analytical methods, protocols and standard reference materials in use [1]. This abstract addresses the comparability of Inductively Coupled Plasma Mass Spectrometry (ICP-MS), Quantitative Neutron Capture Radiography (QNCR), and Prompt Gamma Activation Analysis (PGAA) for the determination of boron in biological samples, using blood and tissue samples from a clinical study for the comparison. The methodical comparison of ICP-MS, ICP-OES, PGAA, and QNCR is of general interest, because these are the most frequently used methods for quantitative analysis in clinically relevant studies

Materials and Methods:
Blood and tissue samples were obtained from a clinical study to investigate the uptake behaviour of p-boronophenylalanine-fructose (BPA-f) in cancerous and tumour free liver tissue. Since the boron distribution within a sample taken from tumour free liver tissue was very homogeneous, it serves as an ideal sample matrix to compare methods for integral and locally selective boron analyses. To be able to compare data from ICP-MS or PGAA directly to those obtained by QNCR, a set of reference blood samples was included in the comparison, which before had been used for the production of reference standards for analysis by QNCR [2]. Irradiations for QNCR and PGAA took place at the research reactors of the University of Mainz, the HFR Petten, the Netherlands, and the FRM II of the Technical University of Munich, Germany.
The larger part of the tissue biopsies could be analysed by all three methods, thus creating a situation similar to a Round-Robin trial. The blood samples were first analysed by PGAA and then by ICP-MS, since the latter required wet-ashing for sample preparation. The measurements by QNCR and PGAA, as well as the related clinical study are described in detail elsewhere [2, 3].
Results and discussion:
Consistency and conformity of ICP-MS and PGAA measurements for blood samples in a range of 0 – 30 ppm ^{10}B was very good. From previous works, it is known that both methods relying on the inductively coupled plasma showed little difference in performance when measuring samples equally prepared following the same

protocol [4, 5]. Also, comparison of ICP-OES to PGAA had revealed good agreement [6].
However, for none of the three methods accordance with QNCR has been found so far. In previous works, Probst et al. concluded that differences to QNCR may be due to the fact that ICP-MS and ICP OES could not account for the very heterogeneous boron concentration that can occur in tissue samples. In this respect, tumour free liver tissue appears as a very suitable “model system” to overcome this problem, which could be demonstrated by very consistent measurements comparing the three methods included in the comparison.

Conclusion:
The data of all methods and their comparison revealed that it is possible to obtain matching results from all three methods for a specific type of organic sample (tumour free liver tissue). also demonstrating the possibility to obtain consistent results from analytical techniques for integral boron determination in comparison to a locally selective method.
Reference materials in use for analytical, and especially radioanalytical, boron determination by groups worldwide vary in their characteristics depending on each method. For standardisation of analytical protocols, BPA-f in whole blood is suitable for comparison of ICP-MS, ICP-OES, PGAA and, indirectly, for QNCR and could therefore serve in a larger Round-Robin trial as reference material.

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ORAL PRESENTATION - On the development of a rapid method for the concentration and separation of radiostrontium from water samples based on a new Sr selective resin

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Due to its radiotoxicity there is a strong need for the determination of Sr-90 in environmental samples. One of the main pathways for Sr-90 ingestion is the consumption of water; accordingly the determination of Sr-90 in water samples is of special importance, in routine analysis for environmental monitoring as well as in emergency situations and contamination control. Rapid methods are of high interest in both of these cases, in emergency situations and contamination control, since fast results are needed in order to evaluate a situation thus allowing appropriate measures to be taken, and in routine analysis as they allow fast sample turn-around time and thus high sample throughput.

In order to allow the quantification of Sr-90 by β-spectrometry it is necessary to perform a number of sample preparation steps upfront to the measurement: sample pre-treatment, chemical separation and the preparation of counting samples. Sr-90 is frequently concentrated from water samples using co-precipitation or ion exchange methods both requiring considerable hands-on and overall lab time.

Some results of the development and characterization of a new Sr selective resin, allowing direct concentration and separation of 90Sr from environmental water samples are presented. A number of candidate resins were prepared based on the Sr selective crown-ether di-t-butyl dicyclohexyl-18-crown-6 and varying amounts of di(2-ethyl-hexyl)phosphoric acid, selected room temperature ionic liquids (RTILs) respectively. Best suited candidate resins were identified, and further characterised, through batch experiments. Based on obtained results a separation scheme has been developed and optimized, allowing Sr to be successfully separated from synthetic samples.

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ORAL PRESENTATION - Separation of Uranium and Polonium in drinking water by calix[6]arene columns

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The general population is chronically exposed to uranium and polonium mainly through day-to-day food and beverage intake. The measurement of these naturally-occurring radionuclides in drinking water is important to assess their health impact. The methods currently used for these analyses require tedious sample preparation techniques (evaporation, precipitation, column separation...) and long counting times by alpha spectrometry. Our laboratory developed calix[6]arene molecules able to entrap the actinides U, Pu and Am from complex matrix like urine samples. In this work, the applicability of calix[6]arene columns for uranium and polonium analysis in drinking water was investigated. The analysis of uranium in water is very simple with calixarene column, named AQUALIX. Drinking water sample (until 500 ml) can be directly loaded onto AQUALIX, without prior specific treatment except an easy step of acidification and gas extraction, to eliminate soluble carbonate species. Then uranium can be eluted in acidic solution from column in order to perform the measurement by inductively coupled plasma mass spectrometry (ICP-MS) or alpha spectrometry. This new procedure is suitable for routine analysis and requires a considerably reduced number of sample treatment steps as compared to usual procedures. In this way, the total analysis time is around half a day by combining the separation on AQUALIX with fast ICP-MS measurement. In case of simultaneous presence of polonium and uranium in water, the separation of both radionuclides is essential due to interferences in spectrometry alpha measurement. So we developed a protocol based on a first step of spontaneous deposition of Po on metallic disk followed by the U extraction on AQUALIX column. This new procedure was applied with success to different French drinking waters (still mineral water, sparkling mineral water, tap water).

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OPENING LECTURE - CINCH - Cooperation in education In Nuclear Chemistry

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The renaissance of nuclear power is already requiring a significant increase in the number of the respective specialists, amongst others are nuclear chemists. The project Cooperation In education in Nuclear CHemistry (CINCH) aims to coordinate the current fragmented and diverse activities in Nuclear Chemistry education and training in Europe both at Ph.D. and undergraduate levels, in collaboration with Russia. The system developed should enable formation of a long-term Euratom Fission Training Scheme (EFTS) and contribute to moving the education and training in nuclear chemistry to a qualitatively new level. The work has been coordinated also with activities of ENEN association, Division of Nuclear and Radiochemistry of EuChemMS, and the training modules of EUROATOM “chemical” IPs and NOEs.

The main results of the project with the broadest impact to students, teachers, industries, and research community will be a set of compact joint modular courses in different branches of modern nuclear chemistry, an e-learning platform available for both education and training (both applicable at the Ph.D., life-long learning, and MSc. levels), and a long term sustainable strategy for nuclear chemistry education. In this presentation, examples of the structures of the courses developed will be given and other project activities will be reviewed.

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INVITED LECTURE - Nuclear and radiochemistry education in European Universities

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Within the EU project CINCH (Cooperation in education in nuclear chemistry) a survey on education of nuclear and radiochemistry (NRC) in European universities was produced by the University of Helsinki, Laboratory of Radiochemistry. Altogether 22 countries and 69 universities were covered by internet survey and by sending a questionnaire to appropriate departments. 42 universities, including most relevant institutions, replied to the questionnaire. The survey was mainly focused on teaching at the master's level which is the typical level of teaching. 83% of the universities give NRC teaching at the MSc level or both at MSc and BSc levels and 17% only at the BSc level. In addition, a number of universities have high quality doctoral programmes in NRC but have no or only limited teaching at lower levels. In general, the teaching programmes at various universities are very diverse.

Only a limited number of universities have specific MSc and BSc programmes for NRC. Only two universities (3% of surveyed universities) have a BSc programme and five universities (7%) an MSc programme. In addition to these, specialization in NRC within chemistry MSc programmes is possible in 25 universities (36%). However, in most of the universities (57%) only a few courses in NRC are taught under various MSc programmes. In the NRC master's programmes and in specializations (44 altogether) most (55%) universities focus on basic NRC while 20% of them focus in environmental radioactivity/radioecology, 16% in nuclear energy and materials and 9% in radiopharmaceutical chemistry.

Most of the courses given (57%) can be categorized to basic NRC which mainly covers basic nuclear physics as well as detection and measurement of radiation. In addition, 14% of the courses deal with environmental radioactivity and radioecology, 11% analytical radiochemistry, 7% nuclear energy and materials 7% radiopharmaceutical chemistry.

Both the survey report and the list of universities giving NRC education have been published at the CINCH home page http://cinch-project.eu/.

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INVITED LECTURE - Nuclear and Radiochemistry Training in the European system of Accumulation and Transfer of Credits for Vocational Education and Training in Europe (ECVET)

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ECVET is based on concepts and processes, which are used in a systematic way to establish a common and user-friendly language for transparency, transfer and recognition of learning outcomes in Europe. The European Parliament and the Council have adopted ECVET in 2009 and since then a number of projects dealing with implementation of ECVET have started.
Eleven projects between 2008-2011 and eight between 2011-2014 respectively have been running, which are funded through the centralized actions of Livelong Learning Programme. A series of other projects, funded by national programmes or other European funding schemes not directly interested in credit transfer are also on-going.
Nuclear and Radiochemistry Training in the ECVET European system is going to be further discussed, with examples. A series of achievements are going to be highlighted, as well as the limitations and opportunities in the present context in nuclear field.

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INVITED LECTURE - Skills and Knowledge Structure Needs - End-users' View

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The CINCH project aims to coordinate education in nuclear chemistry, both at PhD and undergraduate levels, within the EU and Russia; targeting doctoral, masters' students and research workers. Including these students into the system should increase attractiveness of the studies of nuclear chemistry and thus enlarge the source of highly qualified professionals for the future employers.
Across the EU and Russia there is a varied level of training available. In the UK, there exists a well established base for nuclear education available from a wide number of universities, at both undergraduate and post graduate level; funded by the RCUK and privately through student fees. On the whole this follows a 4+4 (MSc/MEng + "long" PhD) version of Bologna. Although some institutions still offer the classic 2 year MSc in nuclear related topics. Due to the large number of courses on offer, the provision of nuclear training is market driven, with students choosing on the basis of attractiveness to industry. Industry is supportive of this approach as it means that they do not have to bear the cost of education.
Across the EU we need to move to similar position, so that industry will able to recruit from a large pool of qualified graduates. The issue facing most EU countries is that there are not enough higher educational institutions capable of delivering nuclear education, and in the case of CINCH nuclear and radiochemistry education, to offer the necessary diversity and numbers of students to satisfy demand. The solution is to form a consortium that offers the full range of subjects required and with each member offering courses that match their speciality. This is the model being developed by CINCH for nuclear and radiochemistry.

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ORAL PRESENTATION - Curriculum Development for Nuclear Fuel Chemistry, Reprocessing and Separation Chemistry, and Radioactive Waste Management at the Pennsylvania State University

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Recent curriculum development in the Department of Mechanical and Nuclear Engineering at Penn State University includes Laboratory Experiments in Applied Nuclear and Radiochemistry and a new Nuclear Security Education Program (NSEP) being develop in collaboration with MIT and TAMU with the support of the DOE-NNSA, Global Threat Reduction Initiatives. To supplement the NSEP and attract more undergraduate students in radiochemistry, development of several new course modules has been started at Penn State with US Nuclear Regulatory Commission funding. The course modules are structured in two tiers, introductory and advanced; students will be required to satisfactorily complete the two introductory modules before enrolling in the advanced modules. The introductory module Introduction to Actinide and Lanthanide Chemistry provides a review of basic nuclear and chemical concepts, as well as more advanced concepts on the electronic structure, aqueous behavior, and solid behavior of the actinide and lanthanide elements. Introduction to the Nuclear Fuel Cycle introduces the most common types of nuclear reactors and the collective process of the nuclear fuel cycle, including fuel harvesting, processing, use, and disposal. Four advanced course modules provide in-depth instruction on specialized areas of the fuel cycle and actinide chemistry. Nuclear Fuel Chemistry presents the chemistry of nuclear fuels in three parts: the characteristics and manufacture of nuclear fuel; alteration processes in nuclear fuel during irradiation; and post-irradiation chemical behaviors of nuclear fuel under storage or disposal conditions. Nuclear Fuel Reprocessing and Separations Chemistry reviews the radionuclide inventories for used commercial fuel bundles of typical burnup levels, as well as the overall objectives and outcomes required of any potential reprocessing method. Detailed studies of the chemistry of several important historical and research-scale reprocessing methods are emphasized. Radioactive Waste Management presents current and potential waste disposal options for all parts of the nuclear fuel cycle. In Environmental Radiochemistry, students are presented with the most important radionuclides of environmental concern and the environmental processes that influence their behavior. Details of the development of radiochemistry course modules at Penn State will be presented.

Panel Discussion: Minimum Requirements for a Master's Degree in Nuclear and Radiochemistry - Towards European Master's Degree / 266

Panel Discussion - ● Introduction: Prof. J. Lehto; ● Panelists - 3 minutes per panel member; ● Discussion: 10 minutes; ● Panel Summary: 5 minutes

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INVITED LECTURE - The Behavior of Actinide Elements in Contaminated Environments

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Savannah River Site (SRS) is one of several US Department of Energy (DOE) sites that have been used to produce nuclear materials for defense, industrial, and medical purpose. During several decades of processing, low-level radioactive effluents were discharged at the site, which have subsequently migrated away from the disposal area. Remediation of the area is needed; however, remedial action requires a thorough understanding of the contaminant behavior in this environment.

In this study, the distribution of contaminants in soils collected from the SRS F-area seepage basin and down gradient from the source was quantified for U, Pu, Am, and Cm using radioanalytical and mass spectrometric techniques. The results suggest that there are two sources of U and three sources of Pu in this area. In addition, using published groundwater data, Pu and Cm behavior in the groundwater is proposed . Also, the partitioning of actinides to these soils was studied using sequential extraction. The results indicate a source dependency for actinide partitioning in the soils. This information can be used by remediation engineers to design appropriate cleanup alternatives for the area.

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INVITED LECTURE - Chemical dosimetry for BNCT mixed radiation field and conformal radiotherapy

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Gel dosimeters allow in-phantom verification of the absorbed dose spatial distribution in conformal radiotherapy treatments. Particularly advantageous are Fricke gel dosimeters in form of layers that can be analysed with very simple instrumentation, give precise results if properly calibrated and offer particular advantages in the mixed neutron-gamma fields of boron neutron capture therapy (BNCT).
Fricke gel layer dosimeters are based on ferrous sulphate solution (Fricke solution) containing Xylenol-Orange, infused in 3mm-thick gel matrix. The measurable effect produced by ionising radiation is a change in the wavelength of visible light absorbance. The dosimeters are imaged with a CCD camera system before and after irradiation, and the measured difference of optical density around 585nm wavelength is proportional to the absorbed dose. The good tissue equivalence of such dosimeters, consisting in a dilute water solution, and the independence of their response on photon energy in the range of interest for radiotherapy, constitute valuable characteristics for appropriate in-phantom dosimetry in conformal radiotherapy. Moreover, the layer geometry has allowed the development of a calibration procedure that enable achieving very high precision (within 3%).
The layer geometry of such gel detectors is particularly convenient also in BNCT dosimetry. In fact, a method for imaging the various dose components has been proposed and widely applied, based on couples of gel-dosimeters having a suitable difference in their isotopic composition. Thanks to layer geometry, neutron transport is not sensibly affected by such a gel matrix variation because it is mainly determined by the tissue-equivalent phantom around dosimeters. Therefore, it is possible to measure the spatial distribution of the dose due to the charged particles generated by B-10 reactions with thermal neutrons, of the gamma dose due to background and to photons emitted in the reactions of thermal neutrons with hydrogen and also of the dose due to the fast component of the epithermal neutron beam.

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INVITED LECTURE - Determination of the isotopic ratio $^{236}U/^{238}U$ in environmental samples

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236U with a half-life of 2.3·107 years is produced via thermal neutron capture on 235U. In nature these neutrons may result from (α,n) reactions on lighter nuclides, spontaneous fission of 238U, induced fission of 235U and at the Earth surface they are part of the cosmic rays. Only small amounts of 235U are produced naturally from uranium in ores, soils and rocks, but a huge amount is produced in nuclear power plants. While naturally 236U occurs in ultra trace concentration in the environment with an expected isotopic ratio of 236U/238U in the order of
10-14 to 10-13(1), nowadays the ratio measured in surface samples is clearly elevated due to the anthropogenic input.
For the analysis of the 236U/238U ratio, water samples from rivers and creeks were collected in the alpine region of Austria, from the Danube, the Black Sea, Irish Sea, and from the Atlantic and Pacific Ocean. From areas in Salzburg and from the clean air site LaPalma Island, Spain, also soil samples were investigated. Retrospective 236U levels were measured in the antlers of red deer. Natural ratios without any anthropogenic contamination were measured in uranium ore and yellow cake samples from Australia, Brasil and Canada.
After a pre-concentration and an anion exchange step the uranium fraction was
co-precipitated with NdF3 and thin sources were prepared for α-spectrometry to determine the activity ratio of 234U/238U and the chemical yield. Afterwards these filters were reprocessed for the analysis of the isotopic ratio 236U/238U by AMS (Accelerator Mass Spectrometry) (2). The special aim of our research was the characterization of the 236U/238U ratio in environmental samples, and to investigate the contribution from anthropogenic sources.

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INVITED LECTURE - Study of neptunium sorption on clay and clay minerals using X-ray absorption spectroscopy

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In several European countries argillaceous rocks have been considered as potential host rock for the construction of radioactive waste disposal repositories. Among potential migration paths, sorption and diffusion of radionuclides are the most important processes for the migration of these elements beyond the engineered barriers of the repository.
We have investigated the sorption of neptunium(V), the most mobile and soluble species of Np, on clay minerals [1] as well as on natural Opalinus clay (Mont Terri, Switzerland) [2]. The interactions between Np(V) and the clay and clay minerals have been studied by batch experiments as a function of several chemical parameters and by synchrotron-based techniques.
Before presenting the results for Opalinus clay, the most important findings of batch and EXAFS studies of Np(V) sorption on pure clay mineral phases will be briefly reviewed. Since Opalinus clay has a heterogeneous composition, the sorption of Np(V) has been studied by microfocused synchrotron X-ray fluorescence (μ-XRF) mapping, microfocused-XANES spectroscopy (μ-XANES) and microfocused X-ray diffraction (μ-XRD) at the Swiss Light Source. By combining these synchrotron-based techniques, the following results were obtained [3]: 1) Considerable amounts of Np(IV) were detected in areas that are enriched in Np, even when the sorption experiments with Np(V) were conducted in air. 2) Under anaerobic conditions, a correlation between Np and Fe was observed in the μ-XRF maps, indicating that the reduction of Np(V) is caused by an iron(II)-containing mineral that could be identified as pyrite by μ-XRD. The reduction of Np(V) to the less mobile Np(IV) is an important process that increases the sorption of this long-lived radiotoxic element and retards its migration through the clay.

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INVITED LECTURE - Trace Element Analysis with high sensitivity spectrometry.

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In high sensitive experiments for rare events physics searches, the determination of contaminants is one of the most critical issue. Radioactive contamination in constructing materials can mimic the tiny signal of the studied events and this reduces the global experimental sensitivities. To fulfill the requests of experiments like double beta decay or dark matter searches, sensitivities in the scale of microBq/kg for 238U and 232Th chains are actually needed: such measurement capabilities are normally not accessible from standard instrumentations.
Many spectroscopic techniques were developed in order to reach the requested sensitivities and to guaranty the correct selection of the detectors materials. In this way also the construction of the spectrometers need particular cares and specific approaches.
Actually HPGe spectrometers specifically constructed reach very high capabilities in the detection of very low radioactive background in ultra pure materials. These detectors were realized using the same approach as that adopted for low background experiments that give us the methodological approach to increase their measurement sensitivities.
Methods, instrumentations and specific data analysis developed for very high sensitive measurements will be presented. Applications of these high sensitivities techniques to other field of scientific researchers will be also discussed.

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ORAL PRESENTATION - Coupling actinide speciation and thermodynamics: Neptunium(VI) solubility and speciation in alkaline NaCl solutions

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In order to assess the long-term safety of a nuclear waste repository it is essential to derive robust predictions of radionuclide solubility and speciation. Based upon correct and reliable chemical models it is possible to derive comprehensive sets of thermodynamic data and quantitatively analyse radionuclide behaviour for different scenarios. Coupling modern actinide speciation tools and conventional thermodynamic approaches offers important advantages for developing comprehensive quantitative geochemical models. The use of advanced spectroscopic tools in actinide chemistry, one main driving force for aqueous actinide chemistry over the last decade, yields detailed molecular level information on chemical speciation and aqueous and solid phase characteristics. Conventional solubility studies allow deriving robust solubility limits and information on macroscopic thermodynamic properties like solubility products, complex formation constants or ion-interaction parameters. Improved spectroscopic information on actinide speciation will consequently result in improved chemical models and thus contribute to improved thermodynamic descriptions and geochemical model predictions.

In recent studies of Np(VI) solubility in alkaline NaCl solutions we have successfully combined advanced spectroscopic studies with information from solubility studies and chemical thermodynamics. Using XAFS-techniques, we were able to derive stability fields for Np(VI) in aqueous solutions and correlate this to data derived from conventional Eh and pH measurements. Having established the relevance of Np(VI) we have proceeded to perform integral solubility studies in dilute to concentrated NaCl solutions over the entire alkaline pH range. The studies show a distinct dependence of the Np(VI) solubility on pH and ionic strength conditions and indicate significant Np(VI) retention over a large set of chemical boundary conditions. By analysing the solubility data and comparing to the previously investigated chemically analogue U(VI) system, we were able to derive a comprehensive thermodynamic model for the system Np(VI)-Na+-H+-OH--Cl--H2O at 25°C not available before.

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ORAL PRESENTATION - Mobilisation of radionuclides and heavy metals from mill tailings at a former uranium mine in south east Finland

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Many new ore prospecting projects have been launched recently in Finland, seeking to exploit both metalliferous and non-metalliferous (e.g. phosphate) mineral deposits. Currently there is increasing awareness of the radiological impact of non-nuclear industries that extract and/or process ores containing naturally occurring radioactive material (NORM). These industrial activities may cause significant environmental problems if the waste generated during processing is not adequately managed.

The Paukkajanvaara mining site in Eno started operation in 1959. The mine was a test site for assessing the feasibility of larger scale uranium extraction. The ore was milled and enriched on site but mining proved to be uneconomic, and so operations ceased in 1961. In the late 1970's the entrance of the mining shaft was sealed with a concrete slab and the area including tailings, waste-water ponds etc., was abandoned. The site was left untouched for nearly 30 years until Finnish Radiation and Nuclear Safety Authority initiated a review. Their results indicated that ambient radiation levels at the site had increased by 0.5 μSv/h as a direct result of the earlier mining operations. Rehabilitation was completed by 1994 after which the area was released for outdoor use without restrictions.

The aim of this study was to examine the potential for mobilisation of radionuclides and heavy metals from the mill tailings and the waste rock pile. Samples of run-off sediment were collected along a transect from the waste rock to a small pond (Iso Hiislampi). Depth-profiled bottom sediment samples were also taken from the pond close to the mill tailings area. After pre-treatment, gamma spectrometry was used for direct determination of uranium and thorium progeny (principally, ^{234}Th (^{238}U), ^{235}U, ^{226}Ra, ^{210}Pb and, - ^{228}Ac (^{228}Th)). Samples were then analysed by X-ray diffraction (XRD) to determine the main minerals present. Thereafter, aqueous samples were prepared by microwave digestion in nitric acid allowing determination of uranium, thorium and heavy metals by Inductively Coupled Plasma Mass Spectrometry (ICP-MS) and uranium and thorium by alpha spectrometry, respectively. For the latter method, uranium and thorium were separated using anion exchange chromatography. The use of complementary analytical techniques highlighted issues with complex mineralogical matrices and reduced uncertainty in the extent of legacy radionuclide and heavy metal contamination present at the site.

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ORAL PRESENTATION - Radioanalytical determinations and radioactivity measurements in the field of Radiation Protection: the experience of the Integrated Laboratory of Radioactivity Measurement and Monitoring of the ENEA Radiation Protection Institute

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Since more than 40 years ENEA has been working in the field of radioactivity measurement especially for environmental and internal dosimetry monitoring purposes. This activity represents a great heritage of competences and experience, now collected in the Integrated Laboratory of Radioactivity Measurement and Monitoring of the ENEA Institute of Radiation Protection (IRP MIR), which acts in three different units located respectively in the ENEA Centers of Saluggia, Casaccia and Trisaia. Thanks to their favorable geographic positions (placed respectively in the North, Center and South of Italy), these units work as a single one Service, able to intervene all over the country.

The large set of measurement systems and equipments available at IRP MIR, together with the knowhow for the application of specific techniques of physical-chemical and radiochemical preparation, allow to carry out analyses on a wide spread of different samples for the determination of a huge amount of radionuclides, particularly those of the greatest radiation protection interest. IRP MIR labs are continuously subjected to internal verification through intercalibration tests and the quality of their performances are supported by regular participation to qualifying exercises and proficiency tests proposed by the most accredited national and international organizations, such as PROCORAD (Association for the Promotion of Quality Control in Radiotoxicological Analysis) to which our Institute has been participating since more than 15 years, and ALMERA (Analytical Laboratories for the Measurement of Environmental Radioactivity), the international network of Laboratories established by the IAEA with the aim to give radioanalytical sustain to the Agency, for measurements on samples coming from contaminated areas, because of accidental or intentional release of radioactivity. On behalf of the Italian Government the Institute of Radiation Protection of ENEA is active member of ALMERA network since 2005.

The aim of this work is to present an overview of the performances provided by IRP MIR Integrated Laboratory in the field of environmental monitoring, radiotoxicology and "special matrices" radioactivity analyses, while giving particular emphasis on the types of analyses where the chemical and radiochemical procedures assume a role of greater significance.

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OPENING LECTURE - Radioanalytical techniques in natural environmental radioactivity determination

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Inexpensive analytical tools such as gamma spectrometric measurements are usually applied in radiological monitoring of contaminated areas. However, detection limits obtained by such methods may not fit for the purpose, in particular when biological materials are to be analysed, so more sensitive techniques have to be considered. Appropriate option is radiochemical determination involving alpha-spectrometric or beta-counting detection, as well as measurement following neutron activation. Radiochemical procedures for the determination of U-238, Ra-226 and Pb-210 are presented and critically compared with the gamma spectrometric measurements, as exemplified by a case study of the Žirovski vrh mine and milling facility in Slovenia.

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INVITED LECTURE - Distribution and transfer of radionuclides including iodine-131 in Japanese environment following the Fukushima nuclear accident

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A large amount of radionuclides was released from the accident of Fukushima Daiichi Nuclear Power Plant in March 2011. We have studied distribution of radiocaesium and radioiodine in the environment and their transfer to agricultural products. Shortly after the accident, high iodine-131 concentrations were observed in leaf vegetables harvested in Fukushima and surrounding Prefectures. The levels of iodine-131 in vegetables decreased markedly in April due to the termination of the release and to its short half-life (8 days), then attention has been paid to radiocaesium (caesium-134 and caesium-137) deposited in soil. Results of the monitoring for agricultural crops harvested during spring to autumn showed that no higher values of radiocaesium were found in most vegetables cultivated in Fukushima Prefecture, excluding the evacuation area. However, values higher than the provisional guideline for radiocaesium (500 Bq/kg) were found in some agricultural products such as bamboo shoots, some fruits (e.g. Japanese citron, persimmon) and new tea leaves. These facts could not be explained by root-uptake pathway. Translocation pathway from leaves and barks should be important to understand the contamination of the other organs in the plants. Higher radiocaesium concentrations (>500 Bq/kg) were observed in some areas, although the most of rice in the same area showed the values markedly lower than 100 Bq/kg. We have studied possible mechanisms for the enhancement of radiocaesium in crops.
Since there are not enough data of iodine-131 to understand its detailed spatial distribution, we have analyzed iodine-129 by AMS to examine whether there is any relationship between these two isotopes in soil. It is interesting to note that a relatively good correlation was found between the concentrations of iodine-131 and iodine-129. This finding suggests the usefulness of iodine-129 analysis for the retrospective evaluation of iodine-131 depositions following the accident.

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INVITED LECTURE - Japanese Green Tea: radioactivity measurements, radiochemical extraction yield determination and some radioprotection considerations.

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The aim of this study, performed on a sample of 2011 (year of the nuclear accident of Fukushima) green tea from the Japanese Prefecture of Shizuoka, was to measure by gamma spectroscopy the natural and artificial radioactivity and the dishomogeneity index of the sample, but also a more in depth investigation of the ratio of the two radionuclides of Cesium and the relative transfer mechanisms of radioactivity to humans under normal conditions of use of the product. To do so we reproduced in laboratory the common domestic preparation of the tea beverage doing 4 different extractions and determining the relative yields of extraction for radiocaesium and natural Potassium from the leaves of tea into the beverage. As a consequence we present analytical data to better assess the total radioactivity effectively ingested by consumers of this product and add some considerations on the food safety limits.

Then we added some basic radioprotection considerations to show some common misconceptions in the evaluation of the compliance of this kind of matrices to the food safety limits set by the Japanese law in respect to the FAO's "Codex Alimentarius" recommendations. Starting from the effectively ingested radioactivity we also show the rough committed relative dose for two categories of public: adult and infants.

Seeing the good results of this work, now our aim is to investigate further and more widely both the concentration of radioactive Cesium and the extraction yields in japanese tea samples, including other prefectures of origin and possibly other types of tea. The goal is to confirm the values of the calculated extraction yields of this batch. It's also important to assess the extraction yields starting from the first year after the one of the nuclear accident because the first year is still affected by both fallout and root uptake of Cesium in vegetables while starting from 2012 only the root uptake pathway will be important.

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INVITED LECTURE - Preservation of Cultural Heritage by Radioanalytical Techniques

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Ancient raw materials are degraded and finally detroyed, mainly by humidity taking up dissolved salts from soil, which are deposited on walls surface breaking plasters and material structure as time goes by. This process is added to environment humidity and temperature changes, but in any case is proportional to porosity materials. This is why, such materials as limestone, sand stone and volcanic stones are most difficult to preserve that feldspar or marble, for example. One easy way to measure the relative porosity of any stuff used in the past as raw material, is to cut samples in the shape of small prisms, to weigh each and to place their basis on a layer about 3 mm deep of a Na-22 (0.01 microcuries/ml)radioactive solution during 5 minutes. Once dry out and conditioned in test tubes or small plastic bags, annihilation gamma rays are detected in a low background, well type 3x3'' NaI(Tl) scintillation detector, during suitable time to accumulate as much counts as possible. Counts per time unit (seconds or minutes) and per weight (grams of stuff) are a relative measurement of the material porosity, which is always proportional to its absorbing power. So, when other pieces of same stuff are cut in similar way and put in contact with a warm solution (60°-80°C)of french gelatin (5%), plus potassium sorbate (2.5%) and sodium benzoate (2.5%), warm solution tends to occupy the small air volumes of pores and channels quicker that if it were cold water. About 3 hours later, at room temperature, when the material looks little thicker and brilliant, it is added a concentrate solution (38%)of formaldehyde, about 10-15% of volume of gelatine employed, in order to get tougher and almost quite insoluble in water gelatin, as well as quite unsuitable to create some organic cultures in it, since potassium sorbate and sodium benzoate are used as food conservatives, and formaldehyde is used to preserve corpses. When these pieces are treated with radioactive solution in the same way that previous ones, detection counts per time and weight units are reduced dramatically by factors at inverse proportion than those obtained with no gelatin. Also, it is a reversible process, since it is possible to dissolve the gelatin just by washing with warm water (60°-80°C)while it resists perfectly washing with cold water (20°-30°C). Based on these laboratory results some prehispanic pieces have been treated in Mexico quite successfully, with no problem at all during 3-8 years. This paper presents these results and proposes to perform the procedure in some other mankind's prides such as Haga Sophia, Xian's warriors, Mithraeum in Sutri, Italy, Borobudur in Java island, Indonesia, Sukhothai, Thailand, Ajanta Caves and Maluti Temple in India, and many others all over the world.

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INVITED LECTURE - Detecting and evaluating minimal traces of radioisotopes in environment and foods

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The radioactivity analysis often requires a very high sensitivity to detect minute traces of both natural and artificial radioisotopes. In many cases, to obtain the required sensitivity is necessary to carry out a concentration of the element to be determined. The measurement of the activity can be done by the alpha or gamma spectrometry according to the type of emission of the radioisotope to be determined. Nowdays are frequently requested results about the concentration of uranium in foods and drinkable water, the contamination by DU in the environment and humans, the concentration of artificial radionuclides in the environment as a result of atmospheric nuclear explosions and nuclear accidents. This kind of analysis requires the adoption of sophisticated techniques to obtain a reliable result.

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ORAL PRESENTATION - Isotopic composition of uranium in aerosol samples collected at 120 km south-southwestern of Fukushima before and after the nuclear power plant accident

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The isotopic composition of uranium (U) in aerosol samples collected before and after the accident of the Fukushima Daiichi nuclear power plant (FDI-NPP), occurred on March 11, 2011, was studied. The aerosol samples were collected on filters by an air sampling system in Tokai, Japan (at 120 km south-southwestern of the FDI-NPP). The filter samples were divided into several parts and U isotopic composition in each filter was analyzed. After ashing the filter samples at a high temperature, the filters were dissolved into acid solutions. Uranium was then chemically separated from the matrix using ion exchange and chromatographic resins. After repeated purification of U, the isotopic ratios were measured by sector field inductively coupled plasma mass spectrometry (SF-ICPMS). The compositions of U in the samples collected after the accident show that non-natural U existed in the aerosol. The results of series measurements of radioactive materials before and after the accident indicate that the radioactive materials released from the FDI-NPP into the atmosphere were transported to the sampling station in Tokai, a place at 120 km distance from the FDI-NPP, within a day.

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ORAL PRESENTATION - Fukushima fallout at Thessaloniki, Greece (40°N) and Milano, Italy (45°N)

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An earthquake of magnitude 9.0 occurred on March 11, 2011 on the Pacific Ocean side of northern Honshu, Japan, followed by a tsunami that struck the east coast of the Tohoku region and caused a serious nuclear accident at the Fukushima Daiichi Nuclear Power Plant. The contaminated air masses were transported across the Pacific towards the North American continent, Europe and Central Asia despite dispersion and washout along the trip. Radionuclides such as ^{131}I, ^{134}Cs, and ^{137}Cs were detected in different locations throughout Europe.

Right after the Fukushima reactor accident, systematic analysis of environmental samples was undertaken in order to detect and quantitatively determine the radionuclides due to the Fukushima fallout in Milano and Northern regions, Italy, by LASA Laboratory of the INFN (Instituto Nationale di Fisica Nucleare) Sez. of Milano and in Thessaloniki, Greece by the Laboratory of Radiation Physics. The concentrations of ^{131}I, ^{134}Cs, and ^{137}Cs mainly in air and water, which control the main way of deposition of the radionuclides in the soil surfaces, vegetables and food chain, were determined. Furthermore analysis of soil, grass and milk samples in both sites of investigation were taken place. The estimated effective doses for population related to the contributions of Fukushima fallout due to different pathways were very far below levels of concern.

This work gives the extent of contamination in Northern Greece and in Northern Italy due to Fukushima fallout and interprets the measured activities at the site of investigation as these resulted from a complicated air mass transport. For the interpretation of activity variations of measured radionuclides, the NOAA HYSPLIT backward trajectories model has been used to assess the transport pattern.

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ORAL PRESENTATION - Plutonium radionuclides in stratospheric and tropospheric air: new evidences from measurements in high altitude aerosols

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Here we report some new experimental results on the content of the long-lived plutonium radionuclides (238Pu, 239,240Pu, 241Pu) in the upper tropospheric and lower stratospheric aerosols. Plutonium radionuclides have been injected into the atmosphere by different processes (ex. nuclear weapon tests (NWT), burn-up of the satellite SNAP-9A, accidents in nuclear facilities, etc.). However, stratospheric component stems mainly from high yield (Megaton) atmospheric nuclear explosions and represents the principal source of global contamination with plutonium (Pu). The aerosol samples were collected periodically since 1970 in the frame of the environmental surveillance programme of Switzerland. Air volumes up to a few thousands cubic meters were filtered through cellulose filters during stratospheric flights using military airplanes.

Our measurements show that Pu radionuclides are present in the stratosphere at higher levels than in the troposphere. The isotope ratios indicate that the main origins of Pu in the stratosphere are the NWTs and the burn-up of the satellite SNAP-9A. The lower content in the troposphere reveals that dry and wet deposition removes efficiently most of the Pu within a few weeks to months. This is not the case for the stratosphere where plutonium has a much longer residence time because of its thermal stratification that separates it from the troposphere. Nevertheless, the analysis of aerosols collected during the passage of the Eyjafjallajökull volcano ash plume in 2010 revealed high levels of Pu in the troposphere, comparable to the values typically observed in the stratosphere. The explosive eruption of this volcano threw volcanic fine grained ash into the stratosphere that quickly mixed with stratospheric aerosols and then may have transported some stratospheric Pu into the troposphere.

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ORAL PRESENTATION - Application of microbeam synchrotron techniques to determine the distribution and speciation of plutonium after uptake by Opalinus Clay

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The synchrotron radiation based techniques X-ray fluorescence (XRF) and X-ray absorption spectroscopy (XANES: X-ray absorption near edge structure) have been used to determine the distribution and the chemical speciation of plutonium (Pu) after sorption and diffusion in Opalinus Clay (OPA, Mont Terri, Switzerland). Thin sections of OPA were contacted with 20 µM Pu(VI)-242 in OPA pore water (pH 7.6, I=0.4 M) under aerobic conditions for at least 3 days. For comparison, a OPA bore core in a diffusion cell was contacted with 20 µM Pu(VI)-242 under the same conditions for 4 weeks. The sorption and diffusion samples were investigated at the MicroXAS beamline at the Swiss Light Source, Paul Scherrer Institut, Switzerland. µ-XRF mapping has been used to determine the elemental distribution of Pu and other elements contained in OPA, e.g., Fe and Ca. Regions of high Pu concentrations were subsequently investigated by µ-XANES to identify the oxidation state of sorbed Pu on the surface of OPA. Further, µ-XRD (X-ray diffraction) was employed to gain knowledge about reactive crystalline mineral phases in the vicinity of Pu enrichments. The results of Pu L3-edge µ-XANES spectra on Pu hot spots showed that Pu(IV) is the dominating species on OPA, i.e., the highly soluble Pu(VI) was retained by OPA in the reduced and less mobile tetravalent oxidation state of Pu. µ-XRD results indicated that Pu is localized on or in the close vicinity of the Fe(II)-bearing mineral siderite and the clay mineral illite. Siderite is one of the redox-active mineral phases of OPA, which determines the speciation on Pu after uptake on OPA.

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